

# **Exergy analysis of the methane and electricity production processes from coal gasification**

**Master Dissertation: Orlando Ramirez  
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## Nomenclature

ASU	Air Separation Unit
$c_p^e$	Mean molar isobaric energy capacity
$h_{fg}$	Latent heat of vaporization
IGCC	Integrated Gasification Combined Cycle
Irr	Irreversibility
n	Number of moles
NCV	Net Calorific Value
PCRC	Pulverized Coal Rankine Cycle
$P_o$	Ambient pressure
R	Universal gas constant
$T_o$	Ambient temperature
SNG	Synthetic Natural Gas
W	Work
WGS	Water gas shift
$X^o$	Chemical exergy
$X^{ph}$	Physical exergy
z	mole fraction
$\delta$	Efficiency defect
$\Phi_{dry}$	Fuel factor
$\Delta H_o$	Enthalpy of reaction

## Introduction

The polygeneration process can convert coal, biomass, petroleum or waste to electricity, chemicals or fuels in a high efficient and environmental friendly way. The gasification has gained value by the need to use low price feedstock like lignite, low rank coals, coke or refinery residues with low pollutants emissions. The most interesting gasification resource in commercial application is coal due to its wide reserves and low prices, therefore in this work coal gasification for the methane and electricity production is evaluated in four chapters to determine the exergoenvironmental performance and the sustainability of the processes.

In the first chapter it was developed the literature review of the polygeneration processes. The exergy analyses in the literature were compiled and the theoretical framework was made for the main process stages.

In the second chapter it was considered the simulation of different types of gasifiers such as fixed bed, fluidized bed and entrained flow reactors. The validation and accuracy of the model was determined and finally it was developed an exergy analysis, considering the environmental performance and capital costs.

In the third and fourth chapters a comparison analysis is presented between conventional and emerging processes. In the third chapter it was compared the indirect and direct methanation processes and in the fourth chapter it was compared the integrated gasification combined cycle (IGCC) with the pulverized coal Rankine cycle (PCRC). For each case the analysis is performed through exergy analysis and taking into account the economics and CO<sub>2</sub> emissions.

The final products selected were electricity and methane, because the first one is the energy in its highest grade of quality and methane is the principal component of natural gas which represents the fuel of the near future since it is the most environmental friendly fossil fuel with high energy content.

## **Problem statement**

Coal is the widest energy source in the world, but it also produces the highest greenhouse emissions when it is burned in chemical and power plants. The main application of this fuel is the direct combustion in a boiler to produce electricity, however through coal gasification; it can be converted to other products like fuels and chemicals. Since the beginning of this century the interest on polygeneration processes rose up due to the need to decrease the oil and gas dependency.

In this work it was considered the production of electricity and methane, comparing the conventional and emerging processes through exergy tools. The conventional process considered for the electricity production is the pulverized coal Rankine cycle (PCRC) and the emerging process is the integrated gasification combined cycle (IGCC). The mature technology for the methane production from coal gasification is the indirect methanation and the emerging technology is the direct methanation.

The exergy analysis is implemented for each case because this method is a suitable approach to identify where the losses occur and how individual components impact the efficiency of the plant. Traditional techniques for the study of the plant performance such as energy balances or criterion of performance are based on the first law of thermodynamics which do not differentiate between the different grades of energy crossing the system boundary and provide no information about internal losses. The exergy analysis is the most suitable method since it takes into account the first and second law of thermodynamics [1].

It is necessary to identify an environmental friendly and competitive way to use coal and coal gasification could represent a suitable option for the energy security of countries with wide coal resources. For this reason, the main question of this work states:

**How much can the emerging technologies increase the exergy performance, the sustainability and decrease the greenhouse emissions of power and fuels production derived from coal?**

## **Objectives**

### **General objective**

Evaluate the exergy performance of the electricity and methane production processes from coal gasification, considering the CO<sub>2</sub> greenhouse emissions and its sustainability.

### **Specific objectives**

Evaluate the exergetical, economical and environmental performance of large scale coal gasifiers.

Quantify the exergy losses in the coal to methane and electricity processes and evaluate the capital costs and the CO<sub>2</sub> emissions with and without carbon capture and sequestration.

Compare the conventional and the emerging technologies for electricity and methane production from coal.

Simulate with Aspen Plus the production of syngas, methane and electricity from coal gasification.



## Methodology

Initially a compilation of the exergy works for polygeneration processes is developed through the Universidad Del Norte Database. The works reviewed considered the production of electricity, fuels and chemicals, analyzed with exergy methods as one of the main tools for the evaluation of the performance.

Subsequently the theoretical framework is performed through a bibliography review for the most important process stages such as the coal gasification, air separation unit, gas clean up, the combined cycle and methanation, including the most important reactions and describing its inherent features. The exergy assumptions and equations are also presented, based on the exergy theory by the author Kotas. In the bibliography revision the performance, costs, technical restrictions of the technologies and information required for the simulation of the process are collected from real plants, demonstrations projects and scientific articles.

Steady state simulations at chemical equilibrium are performed with the software Aspen Plus to predict the properties and composition of the gasifiers. The three most important gasifier types such as the fixed bed, fluidized bed and entrained flow gasifiers are simulated with technical restrictions of the next commercial coal gasifiers: Conoco Phillips, Shell, British Gas Lurgi and KRW technologies. The results are compared to real plants reported in the bibliography to validate the model. The results of the model are used to evaluate the syngas composition produced, the environmental performance, the effect of coal type and to perform an exergy analysis.

In order to collect the data needed for the exergy analysis, the IGCC, the direct and indirect methanation are also simulated with Aspen Plus. The property streams of the pulverized coal Rankine cycle are collected from simulations of the US Department of Energy. The exergy analysis is performed considering only the inlet and outlet streams, hence the stages are considered as black boxes for the exergy evaluation. The exergy calculations are developed only for the most important stages latter mentioned and considered the physical and chemical equations which are programmed with the program EES (Engineering Equations Solver) and Excel.

The exergy analysis is developed for each process individually and then the conventional processes are compared to the corresponding emerging technologies; thus the IGCC is compared with the pulverized coal Rankine cycle and the direct methanation, specifically the hydromethanation is compared with the indirect methanation.

# **1. LITERATURE REVIEW**

## **1.1 Background of exergy analysis works of polygeneration processes from coal gasification and CO<sub>2</sub> capture and storage**

In this section it was made a compilation of the exergy works of polygeneration processes which were classified by the final desired products or as a subcomponent of the polygeneration process. Initially it was considered the electricity production with the integrated gasification combine cycle where it was found most of the works. The next papers comprise the fuels only production, the polygeneration processes and single unit analysis of the gasifiers, air separation unit section and the CO<sub>2</sub> capture and storage. All the works reviewed used exergy analysis as one of the main tools for the evaluation of the performance.

### **1.1.1 Exergy analyses of the integrated gasification combines cycle process**

Exergy analysis of IGCC have been investigated in the past and the works reviewed next consider the electricity as the only principal desired product with no polygeneration cases. Previous exergy studies are focused on several directions as the exergy evaluation of single units on the process performance, process comparisons or technology combinations.

Several works looked at the effect of single units on the process performance which can be seen from authors such as Linwei Ma, 2012, [2] that analyzed the influence of integration of the coal water slurry preheating vaporization technology on the exergy efficiency performance of wet feed IGCC systems with and without carbon capture. He compared ten cases of IGCC systems, including three wet feed IGCC systems with distinct integration modes for this technology. Han, 2009, [3] evaluated exergy performance of the electricity production from coal gasification and char fired natural gas partial reforming. The gasifier considered does not refer to any particular technology but assumes that produces significant amounts of energy content char. Kawabata, 2012, [4] proposed and determined the exergy recuperation when some of the steam produced from the combine cycle HRSG is used as gasifying agent. Additionally compared the plant performance of the pre-combustion and post-combustion CO<sub>2</sub> capture for the proposed and the traditional IGCC plant. De, 2004, [5] made an exergy analysis of an IGCC plant with rankine cycle with a variable number of

steam turbines and heat recovery steam generator sections. The exergy advantages and disadvantages of using variable number of units were quantified. Steinfeld, 2001, [6] evaluated the exergy efficiency and CO<sub>2</sub> mitigation potential of a solar thermal gasification of coal with combined cycle and a fuel cell technology. The exergy and environmental are compared with the process using the conventional steam gasification technology. Ong'iro, 1996, [7] determined the effect of gas turbine firing temperature on the IGCC plant exergetic efficiency and compared the exergy destruction in IGCC with the integrated humid air turbine process.

Other authors focused on the IGCC process comparison between different cases or similar process which generates also electricity. Erlach, 2010, [8] compared the performance of conventional IGCC design with pre-combustion carbon capture by physical absorption with various configurations of chemical looping combustion. Key design parameters are varied, identifying its advantages and disadvantages by the exergy destruction rate. Yang, 2012, [9] studied and compared two IGCC cases with two slurry gasifier types and air preheating system for the combustion chamber. For each process stage, the exergy destruction was assessed. Kunze, 2010, [10] made a comparison between the currently IGCC technology and the ideal case and found a significant potential for further technology optimization. A structure exergy analysis of its subsystems and individual component was performed for both cases. Two individual process has been evaluate by Gnanapragasam, 2009, [11] who assessed and compared the energy, exergy and CO<sub>2</sub> separation performance of an IGCC plant and hydrogen production from coal gasification. The polygeneration case was not evaluated, only the process individually.

On the other hand some studies considered technology combinations to achieve higher exergy efficiency. Odukoya, Dincer and Naterer 2011, [12] investigated the exergetic performance of coal and natural gas co-gasification integrated with a combine cycle and with a solid oxide fuel cell. They determined for a plant configuration the maximum fuel cell power output, combined cycle net work output, combined cycle exergetic efficiency, fuel utilization exergetic efficiency, sustainability index, CO<sub>2</sub> emissions, and exergy destruction. Similar technology combination was made by Lobachyov and Richter, 1996, [13] only for coal gasification and by El-Emam, 2011, [14] who considered the effect of changing the reference temperature and pressure ratio of the component on the exergy destruction for the gasification of two coal types.

Table 1. Exergy Analysis of IGCC.

Author	Year	Exergy analysis contribution	Carbon capture
Single units on the process performance			
Linwei Ma	2012	Water slurry preheating vaporization technology	Yes
Han	2009	Coal gasification with Char fired natural gas partial reforming	No
Kawabata	2012	Pre and post-combustion carbon capture comparison for two plant configurations.	Yes
De	2004	The rankine cycle has a variable number of steam turbines and heat recovery steam generator sections.	No
Steinfeld	2001	Solar thermal gasification of coal	No
Ong'iro	1996	effect of gas turbine firing temperature on the IGCC plant exergetic efficiency and exergy destruction comparison between IGCC and the integrated humid air turbine process	No
Comparison between different cases or similar process			
Erlach	2010	Comparison between the conventional IGCC performance with pre-combustion carbon capture by physical absorption with various configurations of chemical looping combustion.	Yes
Yang	2012	Study of two IGCC cases with two slurry gasifier types and air preheating system for the combustion chamber	No
Kunze	2010	Comparison between IGCC currently technology and the ideal case	Yes
Gnanapragasam	2009	IGCC plant and hydrogen production from coal gasification as individual processes.	Yes
Technology combinations			
Odukoya	2011	Investigation of a cofired coal and natural gas IGCC with a solid oxide fuel cell.	Yes
Richter	1996	Coal gasification with combined cycle and solid oxide fuel cell.	No
El-Emam	2011	Effect of two coal types, reference temperature and ratio of the component on the exergy destruction of IGCC and solid oxide fuel cell technologies.	No

Source: [2], [3], [4], [5], [6], [7], [8], [9], [10], [11], [12], [13], [14]

### 1.1.2 Exergy works of fuel production from coal gasification

In this section exergy analysis of fuel production processes without electricity production is considered. These case studies are not wide studied because for most cases the power production represents a good opportunity to enhance the efficiency of the process.

Steinfeld, 2004, [15] determined the maximum exergy efficiency and CO<sub>2</sub> mitigation potential of three hydrogen production processes using concentrated solar radiation and fossil fuels which are natural gas for the thermal decomposition and the steam-reforming and coal for steam-gasification process. Zhu, 2010, [16] evaluated the exergy and energy performance of the coal to fuel process with a single and two stage slurry coal gasifier. The targets fuels are the Fischer Tropsch synthetic crude and synthetic natural gas.

Table 2. Exergy Analysis of fuels derived from coal

Author	Year	Exergy analysis contribution	Carbon capture
Process performance evaluation			
Steinfeld	2004	Maximum exergy efficiency and CO <sub>2</sub> mitigation of thermal decomposition, steam-reforming and the steam-gasification with solar and fossil energy for hydrogen production.	Yes
Zhu	2010	Exergy and energy performance of coal to liquid and gaseous fuel process with two slurry coal gasifier.	No

Source: [15] [16]

### 1.1.3 Exergy analyses of polygeneration processes from coal gasification

One of the big advantages of coal gasification is that it offers the polygeneration possibility of fuels, chemical and electricity. Previous exergy analyses of these plants are focused on a combination of these final desired products where hydrogen, methanol and electricity are the most studied products.

The exergy process efficiency has been investigated in several direction. Malik, 2012, [17] investigated the hydrogen and electricity production of a zero emission plant from coal and biomass gasification. The plant was simulated with the software Thermoflex and two cases were considered with exergy and energy points of view, the coal only operation and the cogasification of coal and biomass, both with CO<sub>2</sub> capture and sequestration. A similar

work was developed by Chen, 2012, [18] but additionally made a sensibility analysis of hydrogen to electricity ratio on the exergy efficiency and Aspen plus was used for the process simulation. The exergy conversion efficiency of coal to methanol, coke, electricity and heat was studied by Anikeev, 1997, [19]. The author presents a mathematical model of the process which is used to assess the global exergy efficiency of the plant with different coal types and different production amounts. Lou, 2011, [20] used exergy methods in a sustainability assessment of the dimethyl ether and power production from coal and natural gas. He calculated the exergy conversion efficiency, economics, process safety and environmental performance of the coal gasification with natural gas reforming and the co-gasification of both feed stocks. A similar case study was considered by Cocco, 2005, [21] with two coal gasifiers with dry and slurry feeding systems and the exergy losses calculated were at four plant subsystems.

Some authors compared the process exergy efficiency with similar plant configuration. Jin, 2004, [22] evaluated the energy savings of electricity and methanol polygeneration process compared with the plants producing these products individually. The process was simulated with the software Aspen Plus and the results were analyzed through exergy destruction rate and graphical exergy diagrams. A similar case study was developed by Zheng, 2009, [23] but the exergy losses are decomposed into five sub-systems which are chemical reaction processes, heat exchange processes, external exergy losses, turbine/mechanical exergy losses and others. Yuanyuan, 2002, [24] made an analogous work but the subsystems were the gasifier, cleanup unit, synthesis, exhaust heat and combined cycle.

In order to reach higher efficiencies several works propose technology combination with coal gasification. Gao, 2008, [25] studied the technology combination coal gasification with the coal fired and conventional coking process for production of methanol and electricity. The exergy analysis made was on component level based with energy utilization diagram. A similar process combination for the production of methanol, dimethyl ether and dimethyl carbonate was investigated by Li, 2010, [26] with an exergoeconomic analysis. The process was evaluated with a proposed mathematical model of the chemical reaction kinetics and the gasifier and coking process were not considered. Han and Jin, 2010, [27] evaluated exergy performance of a electricity and methanol production from coal gasification and char fired natural gas partial reforming. The exergy destruction rate was calculated at a component level.

Table 3. Exergy Analysis of polygeneration processes

Author	Year	Exergy analysis contribution	Carbon capture
Malik	2012	Hydrogen and electricity production of a zero emission plant from coal and biomass gasification.	Yes
Linwei Ma	2012	Global exergy conversion efficiency of coal to methanol, coke, electricity and heat	Yes
Lou	2011	Global exergy conversion efficiency of the dimethyl ether and power production from coal and natural gas	No
Comparison between similar process			
Jin	2004	Energy savings of power and methanol polygeneration process compared with the plants producing these products individually.	No
Zheng	2009	Exergy losses on power and methanol polygeneration process	No
Yuanyuan	2002	Exergy losses on power and methanol polygeneration process	No
Technology combinations			
Gao	2008	Technology combination with the conventional and coal fired coking process for production of methanol and electricity.	No
Li	2010	Exergoeconomics of a methanol, dimethyl ether and dimethyl carbonate from coal gasification and coke oven gas.	No

Source: [17] [18] [19] [20] [21] [22] [23] [24] [25] [26] [27]

#### 1.1.4 Exergy analyses on Coal Gasification

The works review next considered only the exergy evaluation of coal gasifiers and the air separation unit. Kunze, 2010, [28] developed an exergy analysis of different raw gas cooling concepts and compared the cold gas efficiency of different gasifier designs. The gasifiers analyzed were Shell, Siemens and General Electric technologies through Aspen plus simulation. Janajreh, 2012, [29] calculated and compared the overall plant exergy efficiency of plasma gasification and conventional air gasification of coal, tire, municipal solid waste, algae, treated wood, untreated wood, pine needles and plywood through. The air gasification plant was modeled using the Engineering equation solver while Aspen plus was used for plasma gasification.

The operation condition of coal gasifiers haven been studied by Chen, 2012, [30] who optimized the coal gasification process through an Aspen plus model. The optimization

variables were oxygen to coal ratio, steam to coal ratio and steam temperature and the response variables were the syngas yield, syngas concentration, system exergy efficiency and CO<sub>2</sub> separation rate. Öztürk, 2011, [31] made a sustainability study of coal gasification considering the energy and exergy efficiency, exergy destruction, improvement potential and environmental impact. These parameters were determined for several coal types, operating temperature range from 850 to 1000 C and for an air blown gasifier without steam injection which reach chemical equilibrium.

Coal gasification comparison has been made by Raugei, 2004, [32] comparing the thermodynamic efficiency and environmental sustainability of the syngas production from coal gasification with natural gas off shore extraction, and hydrogen production from steam reforming of natural gas and alkaline electrolysis. Besides the exergy and energy analysis, two other methods were applied; the material flow accounting and energy synthesis.

The coal gasification performance has been also compared with the gasification of other fuels. Prins, 2004, [33] studied the effect of fuel composition on the thermodynamic efficiency of coal and biomass gasification. The exergy losses for both cases were quantified and the optimal gasification temperature for the fuel composition was identified. Dudgeon, 2009, [34] used exergy analysis on gasification for the evaluation of different fuels while comparing the effectiveness of gasifying three fossil fuels which comprises coal, petcoke and orimulsion and three biomass fuels which include distillers dried grains, switch grass and oat hull. In addition an exergy analysis of oxyburn processes including an investigation of flue gas recirculation and a comparison between two different gas separation processes for capturing CO<sub>2</sub> was performed. Anheden, 1998, [35] evaluated a gas turbine with chemical looping combustion of methane and syngas from coal gasification. The two cases are compared with conventional combustion of the same fuels through irreversibility generation rate.

Regarding the air separation unit was investigated by van der Ham, 2010, [36] considering two cryogenic air separation with different number of distillation columns; either two or three. The exergy destruction of the different process parts were calculated and compared for both cases and proposed energy integration with other IGCC components. Cornelissen, 1997, [37] analyzed the exergy performance of a three column configuration air separation unit and liquefaction process. The third column was used for argon purification and the plant can produce gaseous and liquid nitrogen.



Table 4. Exergy Analysis of coal gasification

Author	Year	Exergy analysis contribution
<b>Coal Gasification</b>		
Janajreh	2012	Overall plant exergy efficiency of plasma gasification and conventional air gasification of coal, municipal solid waste, and different biomass types
Kunze	2010	Exergy performance of different raw gas cooling concepts
Öztürk	2011	Coal gasification considering several coal types and operating temperatures
van der Ham	2010	Two air separation unit with different number of distillation columns.
Raugei	2004	Comparison of the syngas production from coal gasification with natural gas off shore extraction, and hydrogen production from steam reforming of natural gas and alkaline electrolysis
Chen	2012	optimized the coal gasification process through an Aspen plus model
<b>Performance comparison with other fuels</b>		
Prins	2004	Effect of fuel composition on the thermodynamic efficiency of coal and biomass gasification
Anheden	1998	Performance of gas turbine with chemical looping combustion of methane and syngas from coal gasification
Dudgeon	2009	Gasifying effectiveness of three fossil fuels and three biomass types, and evaluation of oxyburn processes including flue gas recirculation.

Source: [28] [29] [30] [31] [32] [33] [34] [35] [36] [37]

### 1.1.5 Exergy analysis on CO<sub>2</sub> capture and sequestration

The big concern on CO<sub>2</sub> capture is its high power consumption; therefore authors like Tsutsumi, 2011, [38] proposed a new process to reduce the energy lost on CO-shift conversion and CO<sub>2</sub> chemical absorption processes of precombustion CO<sub>2</sub> separation. The new process was based on the self heat recuperation technology and the process simulator Pro/II was used for the analysis.

Various carbon capture methods suitable to be applied for an IGCC plant for power generation was investigated by Iribarren, 2012, [39] as well as Romeo, 2011 [40]. The technologies examined are post-combustion capture using monoethanolamine, membrane separation, cryogenic fractionation and pressure swing adsorption, precombustion capture through coal gasification, and capture performing conventional oxy-fuel combustion.

Finally Lombardi, 2001, [41] made an exergetic life cycle assessment and a classical environmental life cycle assessment for a post combustion gas turbine, a precombustion IGCC plant and a proposed O<sub>2</sub>/CO<sub>2</sub> cycle. The proposed component burn methane and oxygen producing CO<sub>2</sub> which becomes the cycle working fluid. The CO<sub>2</sub> excess, produced in the combustion, is removed in liquid phase without any additional system.

Table 5. Exergy Analysis of carbon capture and sequestration

Author	Year	Exergy analysis contribution
CO <sub>2</sub> capture		
Tsutsumi	2011	Self heat recuperation technology evaluation.
Iribarren, Romeo	2012, 2011	Various carbon capture methods examined.
Lombardi	2001	Exergetic life cycle assessment and a classical environmental life cycle assessment for a post combustion gas turbine, a precombustion IGCC plant and a proposed O <sub>2</sub> /CO <sub>2</sub> cycle.

Source: [38] [39] [40] [41]

## 1.2 Theoretical framework

### 1.2.1 Description of the methane and electricity production processes

#### Introduction to polygeneration processes

The polygeneration processes consist of three main stages where initially the feedstock is gasified, then the product gases are clean up and finally the syngas can be used to produce several products as shown in figure 1. In this study it was considered only the coal gasification for the production of methane and electricity but the process can be powered with other feedstock like biomass petroleum derivatives or waste to produce also liquid fuels or chemicals like fertilizers, solvents and phenols. The process can achieve low emission; therefore it is considered an environmental friendly way to process coal. Gasification, gas clean up, methanation and gas turbine are described next.

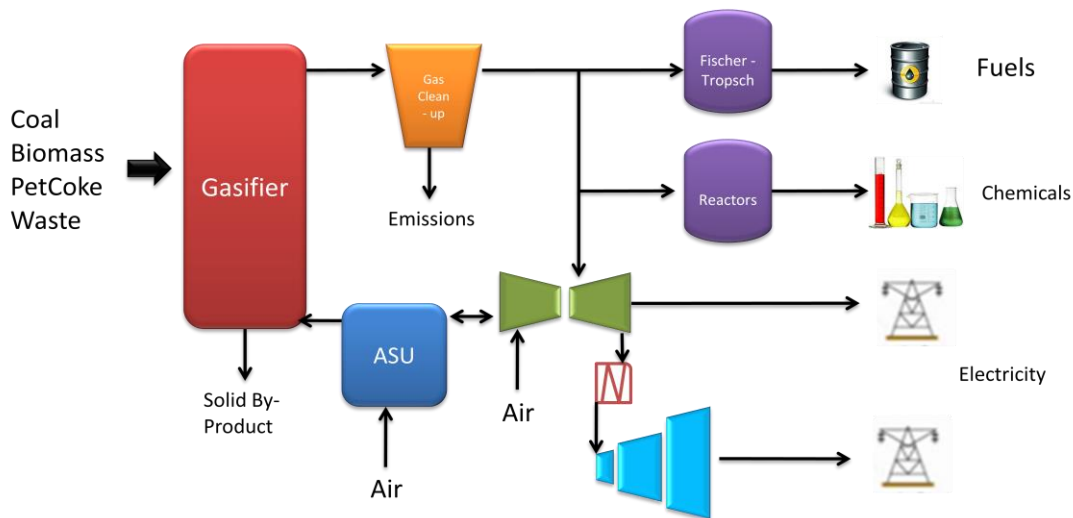


Figure 1. Polygeneration processes. Source: USDoE [42]

#### 1.2.1.1 Coal gasification

##### Coal gasification for the production of low to medium heating value syngas

Coal gasification is the partial oxidation of the components with an oxidation agent that can be air, oxygen, hydrogen, steam or a mixture of oxygen and steam, producing syngas composed mainly of hydrogen and carbon monoxide. Coal gasification with air produces a

gas with low heating value and medium heating value when oxygen is used. The oxidation amount in the reaction should be the gasification stoichiometric amount which is much less than combustion quantity in order to fall in the gasification zone shown in figure 2 and avoiding the complete oxidation, obtaining carbon dioxide and water. Besides carbon monoxide and hydrogen, the syngas produced has a low mole fraction of methane and pollutants as  $\text{H}_2\text{S}$ ,  $\text{NH}_3$  and  $\text{COS}$ . The reactions in the gasifier reach the chemical equilibrium without any catalyst and reach cold gas efficiencies in order of 60 to 90% [43, 44]. The syngas composition depends mainly of the reactor type, the gasification agent and the operational conditions [45].

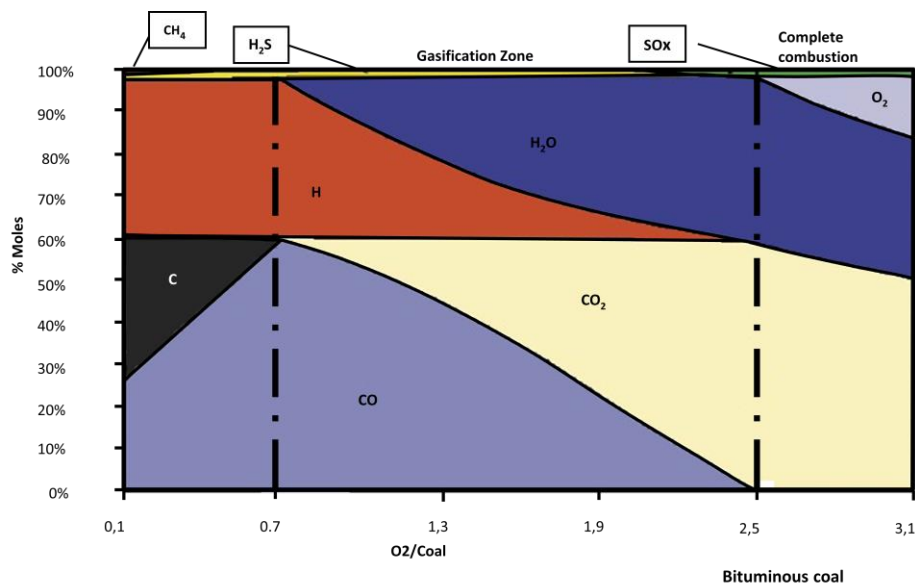


Figure 2. Diagram showing the products of reaction as a function of oxygen to coal ratio. Source: Jeffrey Phillips [43]

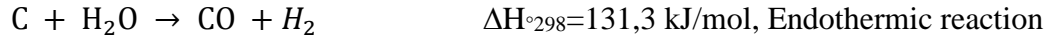
## Reactions in coal gasification

The most important gasification reactions are:

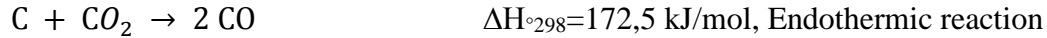
The partial oxidation



The steam gasification



Carbon dioxide gasification



Hydrogasification



Water gas shift reaction



Table 6. Equilibrium constants for gasification reactions. Source: [46]

Log Kp						
T (K)	1	2	3	4	5	6
300	23,93	68,67	15,86	20,81	4,95	8,82
400	19,13	51,54	10,11	13,28	3,17	5,49
500	16,26	41,26	6,63	8,74	2,11	3,43
600	14,34	34,4	4,29	5,72	1,43	2
700	12,96	29,5	2,62	3,58	0,96	0,95
800	11,93	25,83	1,36	1,97	0,61	0,15
900	11,13	22,97	0,37	0,71	0,34	0,49
1000	10,48	20,68	0,42	0,28	0,14	1,01
1100	9,94	18,8	1,06	1,08	0,02	1,43
1200	9,5	17,24	1,6	1,76	0,16	1,79
1300	9,12	15,92	2,06	2,32	0,26	2,1
1400	8,79	14,78	2,44	2,8	0,36	2,36
1	C + 1/2O <sub>2</sub> = CO		4	C + CO <sub>2</sub> = 2 CO		
2	C + O <sub>2</sub> = CO <sub>2</sub>		5	CO + H <sub>2</sub> O = CO <sub>2</sub> + H <sub>2</sub>		
3	C + H <sub>2</sub> O = CO + H <sub>2</sub>		6	C + 2 H <sub>2</sub> = CH <sub>4</sub>		

Table 6 shows the equilibrium constants for each reaction showing that the partial oxidation has the largest participation in the final products and the water gas shift has the lowest.

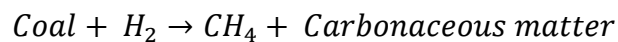
Therefore the oxygen to coal ratio is the most important parameter in order to operate under the gasification zone reaching high carbon conversion and avoiding combustion products like carbon dioxide and water. The steam to coal ratio is another important parameter and the excess of it also hurts the thermal efficiency of the process.

### **Coal gasification to produce high heating value syngas**

These processes produce directly methane in the gasifier. There are two process under investigation; the hydrogasification which gasifies the coal with hydrogen and the steam catalyst gasification, also called the hydromethanation, that uses catalysts to increase the small amounts of methane in coal gasification with steam [46].

### **Hydrogasification**

The process gasifies the coal directly with hydrogen at temperature around the 800 C and high pressure (30-50 bars) in an entrained flow reactors combining the carbon and hydrogen molecules to produce methane without any catalyst [45, 47] . When hydrogen is injected additionally with steam, it enhances the reaction rates, lower the residence time and the coal has not to be dried [48]. After the main reactor, the gas is cleaned and the methane is separated from the unreacted hydrogen and carbon monoxide [45, 47]. The hydrogen must be supplied with an additional unit that can be by electrolysis or with a water gas shift reactor. The most important reaction is:

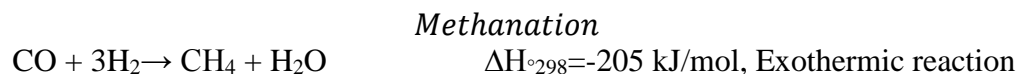
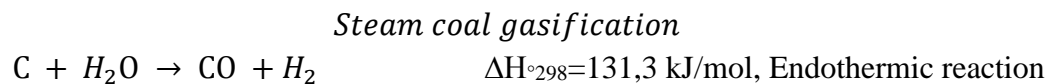


The reaction is exothermic and thermodynamically favored at low temperatures , however a limit temperature is needed to increased its kinetics.

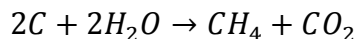
### **Hydromethanation**

In the hydromethanation process, the gasification, water gas shift reaction and methanation take place in the same fluidized bed reactor with the presence of a potassium carbonate (K<sub>2</sub>CO<sub>3</sub>) catalyst at relative low temperatures (600-700 C). The gas yield contains methane, carbon dioxide, carbon monoxide and hydrogen, which are separated with amines and cryogenic distillation and then the unreacted fuels (CO, H<sub>2</sub>) are recirculated in the reactor [45]. The coal is gasified mainly with steam and small amounts of oxygen and the three most important reactions are shown below.

Individual reactions:



Overall reaction:



#### **1.2.1.2 Air Separation Unit (ASU)**

The methods to separate the oxygen and nitrogen from air can be classified in cryogenic plants or non-cryogenic plants. The first one uses a distillation column to produce high purity products at medium to high production rates, therefore the cryogenic option is preferred for polygeneration processes. The non cryogenic plants are membranes technologies or selective adsorption, commonly used when product demand is relatively low and high purity streams are not needed. In this study all the air separation unit (ASU) refers to cryogenic distillation. The ASU produces 95 mole percent oxygen for the IGCC process and for coal to fuels processes the 99 percent separation degree is preferred.

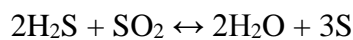
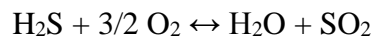
In power generation applications air from the turbine compressor is sent to the air separation unit what is called the integration degree and enhances the efficiency of the process. The separated nitrogen in the ASU is send to the gas turbine, thus increasing the power output while maintaining optimum firing temperature and reducing the NO<sub>x</sub>

formation by dilution. The syngas dilution ranges from 4,4-4,7 MJ/Nm<sup>3</sup> and the specific value depends of the combustion turbine technology.

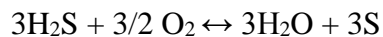
#### **1.2.1.3 Gas clean up**

After the gasification or water gas shift reactors, the gas has to be cooled to 100 C with water quench or heat recovery and the tar and dust have to be removed with syngas scrubber. Then the syngas passes through the acid gas cleanup which can be either the Selexol or Rectisol process, both based on physical absorption [47]. The Selexol process uses a mixture of dimethyl ethers of polyethylene glycol as an absorbent and the Rectisol uses cold methanol and both processes have wide commercial experience. Besides the physical absorption, the clean up process can also use the amine process based on chemical absorption but it is more expensive. The clean up stage separates the H<sub>2</sub>S and CO<sub>2</sub> in two separate streams, which the H<sub>2</sub>S is send to the Claus plant to produce high purity sulfur implementing these reactions:

Individual reactions:



Overall reaction:



#### **1.2.1.4 Water Gas shift reactors (WGS)**

In case of carbon capture or fuels production, the hydrogen fraction in the syngas has to be increased in the water gas shift reaction. The syngas produced in the gasifier is mixed with water and with the presence of a catalyst, the following reaction takes place;

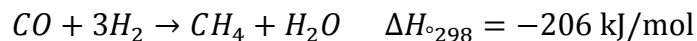




The CO shift converter can be located either upstream of the AGR or immediately downstream. The upstream option is preferred since the water gas shift reaction stage operates at higher temperatures than the gas clean up. In the downstream option the gas has to be cooled, cleaned and additionally steam generated with an additional equipment and energy input.

#### **1.2.1.5 Methanation**

The syngas produced is reformed to CH<sub>4</sub> in the methanation stage with three reactor types; equilibrium-limited fixed bed reactors in series, through wall-cooled fixed bed reactor and the fluidized bed reactor. Through wall-cooled fixed bed reactor and the fluidized bed have been developed only at pilot plant scale and only equilibrium-limited fixed bed reactors are available at the commercial stage [45, 49]. The main reaction in the methanation stage is;



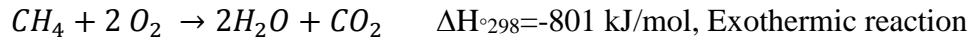
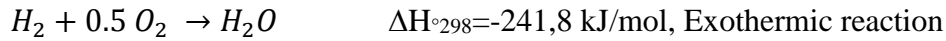
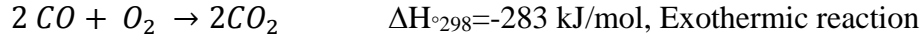
The principal objective on the methanation design process is able to remove efficiently the heat generated by the high exothermic reaction, trying to produce more methane before the chemical equilibrium is reached [50] and to avoid the catalyst sintering and decomposition of the methane to carbon [49]. For this reason several configurations with intercooling and gas recycle were developed with catalysts system that can maintain its activity after prolonged exposure to high temperatures [49]. The main commercial processes are the TREMP and the Johnson Matthey process but reported data in the literature is only available for the TREMP case, which is evaluated in this study.

#### **1.2.1.6 Gas turbine with different fuel composition**

Thermodynamically with a higher turbine inlet temperature, higher will be the efficiency, however the turbine firing temperature is limited to metallurgy restriction. Most of the working fluid is air but the fuel composition also plays an important parameter in the heat transfer to the turbine blades. The gas turbine in this study is an F class gas turbine with a suitable firing temperature with natural gas of 1371 C. For an equivalent turbine lifetime, with a syngas composition of H<sub>2</sub>/CO ratio equal to 0.5, decreases to 1338 C because the heating value of the syngas is lower and more fuel has to be injected what increases the heat transfer by convection. Furthermore with a higher hydrogen fraction also the firing

temperature has to decrease because the water content in the product gases increases and this compound intensify the heat transfer by radiation and convection to the blades.

The most important reactions taking place in the gas turbine combustion chamber are:



The integration between the air separation unit and the turbine compressor is recommended to enhance the global efficiency. The recommended integration degree is 25 to 30 percent of the ASU air coming from the turbine what provides the best balance of plant output, availability, efficiency and reliability [51, 52]. However for CO<sub>2</sub> capture case, all of the available combustion air is required in the gas turbine to maintain a high performance.

## 1.3 Exergy Theory and General Assumptions

### 1.3.1 Exergy assumption

In this work only the physical and chemical exergy were considered and the rest exergy terms as the mechanical, electric, magnetic, nuclear, potential and kinetic were neglected without significant different results. The physical exergy ( $X^{\text{ph}}$ ) considers the maximum work from the temperature and pressure and the Chemical exergy ( $X^{\text{o}}$ ) considers the maximum work from chemical reactions. The chemical species considered in the study are argon (Ar), methane (CH<sub>4</sub>), carbon monoxide (CO), carbon dioxide (CO<sub>2</sub>), carbonyl sulfide (COS), hydrogen (H<sub>2</sub>), water (H<sub>2</sub>O), hydrogen sulfide (H<sub>2</sub>S), nitrogen (N<sub>2</sub>) and ammonia (NH<sub>3</sub>). The ambient condition in the analysis are T=25 °C and P=101.32 kPa.

### 1.3.2 Theoretical exergy models

The exergy models used in this paper are based on the book exergy method of thermal plant analysis by Kotas [1]. The exergy balance at steady state forms the following equations:

### *Exergy of a solid fuel*

The chemical exergy of the coal is described as:

$$x_{coal}^o = [(NCV)^o + w h_{fg}] \varphi_{dry} + [x_s^o - (NCV)_s^o] s$$

For rhombic sulfur:

$$[x_s^o - (NCV)_s^o] = 9417 \frac{kJ}{kg}$$

$$\varphi_{dry} = 1,0437 + 0,1882 \left( \frac{h}{c} \right) + 0,061 \left( \frac{o}{c} \right) + 0,0404 \left( \frac{n}{c} \right)$$

Where h, c, o, n, s, w are the mass fraction of hydrogen, carbon, oxygen, nitrogen, sulfur and water respectively.

### *Exergy of a chemical reactor*

$$X_{in} + X_{out} - Irr = 0$$

The exergy for a stream is the sum of the chemical and the physic term.

$$X = X^o + X^{ph}$$

$$X = n x_T^o + \sum_k n_k x_T^{ph}$$

The specific chemical exergy of a gas mixture is:

$$x^o = \sum_k z_k x_k^o + R T_o \sum_k z_k \ln z_k$$

The physic exergy of a gas mixture is:

$$x^{ph} = \sum_k n_k x_T^{ph}$$

$$x^{ph} = (T - T_o) \sum_k z_k c_{p,k}^e + R T_o \ln\left(\frac{P}{P_o}\right)$$

*Exergy Balance of a distillation column*

$$X_{in} + X_{out} - Irr = 0$$

$$Irr = W_{in} + X_F - X_T - X_B$$

Air at atmospheric conditions and the nitrogen is vented to the ambient:  $X_F=0$ ,  $X_T = 0$

$$Irr = W_{in} - X_T$$

$$X_T = n x_T^o + n x_T^{ph}$$

$$Irr = W_{in} - n x_T^o - n x_T^{ph}$$

*Criteria of performance*

The efficiency defect is defined by Kotas [1] as the irreversibility of the stage divided by the total exergy input to the process. When it is multiplied by 100, it gives us the percentage of coal exergy lost for each stage which is the criteria of performance used in this work.

$$\% \text{ Exergy lost} = 100 * \delta = 100 \frac{Irr}{X_{in}}$$

### **1.3.2.1 Economic Assumption**

The plant capital costs were reported by the US department of energy with an expected accuracy of -15% to -30% on the low side and a +20 to +50% on the high side [53, 54, 51]. These costs correspond to the total amount needed for the plant construction including the equipment, labor, additional construction materials, taxes and contingencies. Financial costs and first consumable costs were not considered.

## 2 SIMULATION AND EXERGY COMPARISON OF COAL GASIFIERS

### Introduction to coal gasification

Gasification has been a reliably commercial scale technology for more than 75 years and currently exist approximately 150 large scale plants worldwide with more than 430 gasifiers where 50% of the total syngas is derived from coal [55] [56]. The gasifier is the most influent plant stage and its selection sets the main performance and costs [57]. The main commercial gasification technologies able to process more than 1000 tons coal per day are: British Gas Lurgi, General Electric initially developed by Texaco, Shell and ConocoPhillips E-gas initially developed by Dow [56] [43]. Figure 3 shows the total installed capacity for these gasifiers, where it can be observed that the three technologies with the highest syngas production are Shell at the first stage, followed by Lurgi and General Electric. The KRW gasifier is also analyzed to include the analysis of fluidized bed gasifiers, but the General electric gasifier was excluded since there is no enough available information to the analysis.

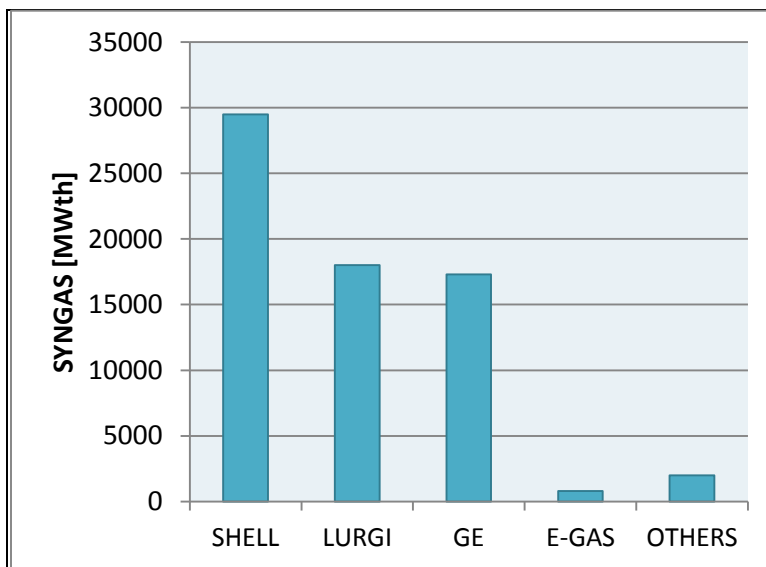


Figure 3 Syngas production capacity from different gasification technologies. Source: [56]

## **2.1 Simulation of coal gasification**

### **2.1.1 Key assumptions**

#### **Description of the gasifier design features**

The gasification technologies considered are a fixed bed gasifier; BGL, a fluidized bed; KRW and two entrained flow gasifier; Shell and E-Gas. The different design options are classified under single stages what determines the features of each gasifier, summarized in table 7 which are considered as inputs and technical restrictions in the coal gasification simulations. The information was compiled from the manufacturer and the US Department of Energy.

The major species considered in the reactor are CO, CO<sub>2</sub>, H<sub>2</sub>, H<sub>2</sub>O, N<sub>2</sub>, O<sub>2</sub> and the minor species tracked are H<sub>2</sub>S, COS and NH<sub>3</sub>. Other low percentage species like mercaptans, metals (Hg, Se) and alkali components were not considered to enhance the model convergence [28].

The coal handling capacity shown in table 7 are for bituminous coal and this value increases for lower rank coals but the energy input capacity decreases. For example the Shell gasifier energy input is 757 MW or 2500 tons/day of bituminous coal and change to 735 MW, 2484 tons/day of subbituminous coal and to 714 MW, 2439/day tons of lignite [54]. The fixed bed and fluidized bed reactor have the lowest coal handling capacity due to these reactors have a residence time on the order of minutes while in the entrained flow gasifiers is on the order of seconds. The coal conversion for all the gasifiers is 99% carbon conversion.

The wall of the gasifier could be refractory lined or use a cooling screen that is a heat exchanger tubes wall that circulates water around the gasifiers to control the reactor temperature and producing low pressure steam. The heat loss for gasifier with refractory lined walls can be neglected but for cooling screen walls with a value in the range of 1 and 2 percent of the heat combustion of the coal feed [58] [59]. Therefore the refractory lined walls reactors were simulated as adiabatic and the cooling screen walls are simulated with a heat loss of 1 percent coal low heating value.

The drying system burns some syngas to heat air which dry the coal to 5 percent moisture content. The drying process requires 25 kg syngas for each ton of bituminous Illinois No. 6 coal.

In the syngas cooling section all the gasifiers use heat recovery steam generator except for the KRW which uses water quench. The temperature of the syngas is cooled to 200 C and

the temperature of the water in the heat recovery was assumed as 280 in the inlet and 335 in the outlet of the heat exchanger at a operating pressure of 15 MPa.

Table 7. Principal design characteristics of coal gasifiers, Sources: [53, 54, 51, 60, 61]

<b>Gasification technology</b>	<b>ConocoPhillips E-Gas</b>	<b>Shell</b>	<b>KRW</b>	<b>British Gas Lurgi</b>
<b>H<sub>2</sub>O/C (mol)<sup>+</sup></b>	0,45	0,11	0,19	0,32
<b>O<sub>2</sub>/C (mol)<sup>+</sup></b>	0,42	0,41	0,48	0,29
<b>Transport gas</b>	H <sub>2</sub> O	N <sub>2</sub>	N <sub>2</sub>	Syngas
<b>Coal drying</b>	No	Yes (5 % Moisture in coal)	Yes (5 % Moisture in coal)	No (Mixed with limestone)
<b>Feeding system</b>	Coal-Water Slurry	Dry coal with lock hopper and Pneumatic conveying with nitrogen	Dry coal with lock hopper and Pneumatic conveying with carbon dioxide	Dry coal with lock hopper and Pneumatic conveying with syngas
<b>Reactor Type</b>	Entrained flow slagging gasifier	Entrained flow slagging gasifier	Fluidized Bed gasifier	Fixed bed slagging gasifier
<b>Gasifier Wall</b>	Refractory	Cooling screen (2 % coal LHV loss)	Refractory	Refractory
<b>Operating Pressure</b>	5,1 MPa	4,2 MPa	3,1 MPa	3,44 MPa
<b>Syngas Outlet Temperature (After Cooling)</b>	1030 C (200C)	1400 C (200C)	1400 C (200C)	537 C (200C)
<b>Gasification temperature</b>	1316 -1427 C	1427 C	1499 C	1400 C
<b>Coal handling capacity (ton/day)<sup>+</sup></b>	2500	2500	880	1000
<b>Syngas Coolers</b>	HRSG	HRSG	Water Quench	HRSG
<b>Pressure loss* (MPa)</b>	1,03	1,07	0,59	1,15

\*Includes the syngas cooling system

+Bituminous coal: Illinois No. 6

## Physical property method

In order to select the proper method, the streams composition and the operating condition of the process has to be considered [62]. The process considered in this study comprises mainly short chain hydrocarbons and non polar substances at high pressure and for this reasons equations of states as Peng Robinson or Redlich Kwong Soave are recommended. In the simulations of this study the Peng Robinson Equation was used as the global property method since it is recommended from Aspen Plus [63].

## Description of the model flow sheet

### *Gasification*

The model flow sheet consists of two reactor and a separator which altogether represents a gasifier. The diagram is shown in figure 4, where the small reactor is a yield reactor. This unit decompose the solid stream into the single components from the proximate and ultimate analysis of the coal and calculates the heating value of the coal which is a input to the second reactor. The second reactor is a Gibbs reactor where the chemical equilibrium from all the streams is calculated and the reactions implemented are shown in table 8. Finally the last unit is a "SSplit" where the ash and the syngas produced are separated.

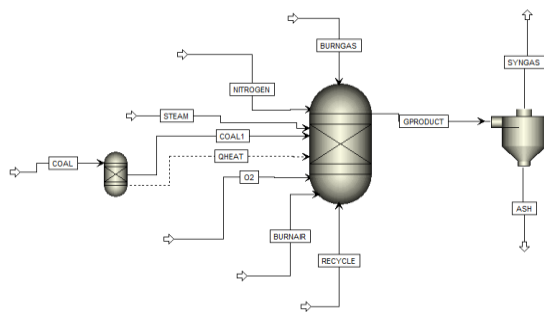


Figure 4. Gasification flow sheet

The second reactor can be a Gibbs reactor because the kinetics barriers associated with its chemical reaction are largely minimized when the temperature are above 873 K, [44, 28] and oxygen steam gasification take place at temperatures higher than 1000 C to certify a high carbon conversion [64] [43]. The chemical equilibrium is calculated from a known



thermodynamic condition when the system reaches the maximum entropy production or equivalent the Gibbs free energy reach a minimum, satisfying the following equation [65]:

$$dG \leq 0 \quad \text{with } (P, T) \text{ constants}$$

Table 8. Reactions taking place in the gasification Gibbs reactor

Stoichiometry	Reaction	$\Delta H_{298}$ (kJ/mol)
$C + O_2 \rightarrow CO_2$	Partial oxidation	-393,5
$C + 0.5 O_2 \rightarrow CO$		-111,4
$C + H_2O \rightarrow CO + H_2$	Steam gasification	131,3
$C + CO_2 \rightarrow 2 CO$	Carbon dioxide gasification	172,5
$C + 2H_2 \rightarrow CH_4$	Hydrogasification	-74,8
$CO + H_2O \rightarrow CO_2 + H_2$	Water gas shift reaction	-41,2
$CO + 3H_2 \rightarrow CH_4 + H_2O$	Methanation	-206.2
$H_2 + S \rightarrow H_2S$	Sulfur reaction	-20.5
$CO + S \rightarrow COS$		-27.9
$CL_2 + H_2 \rightarrow 2HCl$	Hydrogen chlorine reaction	-184.6
$N_2 + 3NH_2 \rightarrow 2NH_3$	Ammonium formation	-91.9

Source: [28]

### *Air Separation Unit and compressors*

Initially the air is compressed to 900 kPa and send to heat exchangers which decreases the temperature to -174 C. The refrigeration performance was taken from the literature. In the column the oxygen is separated to 99 percent purity and then is compressed again to the operating pressure of the gasifier.

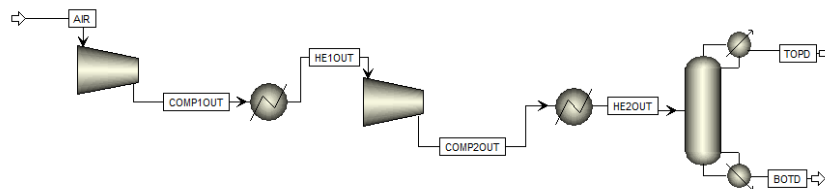


Figure 5. ASU flow sheet

Table 9. Inputs to the model

Global simulation parameters	
Oxygen purity	99%
Carbon conversion	99%
Physical property method	Peng Robinson
Kinetics model	Chemical Equilibrium

### Coal properties considerations

The coal properties have to be considered since it affect the performance of coal gasifiers. The coal type is described in the simulation with the proximate, ultimate and sulfur analysis shown in table 10. Coals are preferred for all gasifiers with low ash content because it decreases the efficiency of the gasifier as a result of an increase in oxygen consumption. Although some gasifiers have a minimum ash content requirement, for example for Shell is >8 wt% because of the slag self coating system on the walls of the gasifiers and to minimize the heat lost [66] [67].

For gasifiers that use mixtures of coal and water such as the E-Gas, the slurryability is an important coal property which is related directly to the coal grindability measured by the hard grove grindability Index (HGI). The HGI shows a size distribution from grindability operations and a high HGI favors the production of slurry with high coal concentration, but it has to be considered if this slurry is pumpable and stable with particle settling [67]. In the simulation a coal water slurry of 63 percent solid concentration is implemented for bituminous and sub bituminous coals.

For gasifiers with low to moderate average temperatures of 1000 °C, reactive coals are desired to achieve high carbon conversion and improving the cold gas efficiency [67]. The gasifiers considered on this work have high gasification temperature of more than 1200 °C and reach high carbon conversion for unreactive coals [68].

In slagging gasifiers the reactor operates above the ash melting point, hence its fusion temperature should be low or moderate so that the molten slag can flow down the reactor walls and drain from the gasifier. Fluxes like limestone can be added to reduce the ash fusion temperature and a temperature of 1400 °C or higher need to consider flux addition. [66] [67]. The software do not considered the ash composition, slag properties and reactivity of the char, hence it is supposed that the slag presents a Newtonian flow, with a viscosity with no crystalline behavior which can blockage the system.

Table 10. Proximate and Ultimate analysis of Bituminous Illinois # 6, Source: [69]

Coal: Bituminous Illinois # 6		
Proximate Analysis (weight %)		
	As Received	Dry
Moisture	11,12	0
Ash	9,7	10,9
Volatile Matter	34,9	39,3
Fixed Carbon	44,1	49,7
<b>Total</b>	<b>100</b>	<b>100</b>
Sulfur	2,51	2,82
HHV, kJ/kg	27,1	30,506
LHV, kJ/kg	26,1	29,544
Ultimate Analysis (weight %)		
	As Received	Dry
Moisture	11,12	0
Carbon	63,75	71,72
Hydrogen	4,5	5,06
Nitrogen	1,25	1,41
Chlorine	0,29	0,33
Sulfur	2,51	2,82
Ash	9,7	10,91
Oxygen	6,88	7,75
Total	100	100

## 2.2 Results

### 2.2.1 Model verification and accuracy

Table 11. Model verification for different gasifier technologies (mol %)

Entrained Flow Reactors					
Dry feed gasifiers			Slurry feed gasifiers		
	Shell			E-GAS	
	Simulation	Reference [28]		Simulation	Reference [70]
CO	60,99	59,27	CO	46,61	44
H <sub>2</sub>	21,65	21,4	H <sub>2</sub>	36,43	33
CH <sub>4</sub>	0	0	CH <sub>4</sub>	3,38	2
CO <sub>2</sub>	1,67	2,8	CO <sub>2</sub>	13,58	16
N <sub>2</sub>	14,07	14,3			
AR	0,96	0,9			
H <sub>2</sub> S	1,27	0,83			
H <sub>3</sub> N	0	0			
COS	0,14	0,31			
Fluidized bed Reactors			Fixed bed Reactors		
	KRW			BGL	
	Simulation	Reference [71]		Simulation	Reference [72]
CO	23,2	23,9	CO	44,66	54,96
H <sub>2</sub>	14,27	14,5	H <sub>2</sub>	35,19	31,54
CH <sub>4</sub>	0,41	1,4	CH <sub>4</sub>	5,33	4,54
CO <sub>2</sub>	5,02	5,5	CO <sub>2</sub>	9,78	3,46
N <sub>2</sub>	51,28	48,6	N <sub>2</sub>	3,95	3,35
AR	0,61	0,6	AR	0,01	0
H <sub>2</sub> O	5,21	5,5	H <sub>2</sub> O	0,96	1,31
			H <sub>2</sub> S	0,03	0,36

The model verification was performed for different gasifier technologies with syngas reported from existing coal gasification plants which are mainly demonstration project. The E-GAS and KWR gasifier projects reported the syngas composition after the gas clean up stage, consequently the accuracy of the model predicting the pollutant emissions for this gasifier was not determined. From table 11 it can be observed that the maximum difference between the Aspen plus equilibrium models and the experimental results reported is 10 percent for the BGL fixed bed gasifier. The most accurate results is presented for the fluidized bed and dry feed entrained flow gasifiers with less than 2 percent deviation for each chemical component, followed by the slurry feed entrained flow gasifiers with less than 4 percent deviation.

The fixed bed reactor produces hydrocarbons liquids such as tars and oils which are not considered by the model. In the BGL gasifier the 7,6 percent of the mass flow corresponds to the hydrocarbons liquids and this is the main reason for the divergence between the simulation and the reference.

Additionally in steady state simulations the gasifier is a perfect mixed reactor with uniform thermo chemical properties where the hydrodynamic within the reactor is neglected consequently geometry is not considered [44]. The BGL is the only technology with a significant different properties along the gasifier which change the equilibrium composition of the syngas. For this gasifier computational fluid dynamics (CFD) has to be implemented to considerer the kinetics within the reactor.

### **2.2.2 Syngas composition and environmental performance**

The slurry gasifier produces a syngas with the highest  $H_2/CO$  ratio approximately equal to 1 which enhances the plant efficiency for the methane production since the downstream methanation needs a  $H_2/CO$  ratio of 3. In case that the syngas has low hydrogen percentage, the carbon dioxide production increases in the WGS what reduces the global plant efficiency. However is has to be considered that the slurry technology presents the highest  $CO_2$  molar fraction, as a result of this gasifier has a high oxygen consumption since more coal has to be burned to evaporate the fluid.

The production of  $CO_2$  greenhouse emissions and sulfur pollutant emissions are presented in table 12 for bituminous coal gasification. The dry feed entrained flow reactor presents the lowest greenhouse emissions, 14 times less than the slurry feed gasifier.

In carbon capture application, the KRW, Shell and BGL syngas have high nitrogen content and it produce a negative effect since it dilutes the raw gas and therefore reduces the  $CO_2$  partial pressure [64]. Therefore the coal compression with  $CO_2$  is preferred for carbon capture cases. The methane production is low for the entrained flow gasifiers as a result of the short residence time, on the other hand the BGL gasifier, with the highest residence time has a significant  $CH_4$  percentage and also the syngas with the highest heating value. The sulfur emissions are also the highest for the fixed bed gasifier [73] and is the only gasifier that produces hydrocarbon liquids, such as tars and oils which need to be recirculated to achieve low pollutants emissions [43]. The 99 percent of the  $H_2S$  can be recovered and send to a Claus plant for the production of pure sulfur [53].

Table 12. Syngas composition for coal gasifier with water quench

	KRW	Shell	E-GAS	BGL
Environmental Performance				
CO <sub>2</sub> , kg/tons coal	186	55	716	153
COS, kg/tons coal	1,5	3	2	3
H <sub>2</sub> S , kg/tons coal	24	25	26	32

### 2.2.3 Exergy analysis

The exergy results are presented in table 13, considering the gasification process as four individual stages; drying, air separation unit, reactor and syngas coolers.

Higher oxygen requirements produce a gas with a lower energy outlet as a result of the reactants are more oxidize. Therefore BGL gasifier counts with the lowest oxygen consumption and the highest exergy conversion efficiency of 82,2%, considering that the liquid hydrocarbons produced are recirculated. The dry feed entrained flow gasifiers are at the second stage in a exergy conversion efficiency with 80,8 percent, followed by the slurry feed entrained flow gasifier with 75,6 percent. The fluidized bed gasifier is in the last stage with a conversion efficiency of 70,4 percent, because this gasifier is recommended only with water quench, destroying 9,1 percent of its exergy. On the other hand Shell recovers 51,5 MW of the exergy cooling the syngas and destroys only 2,1 percent of the exergy.

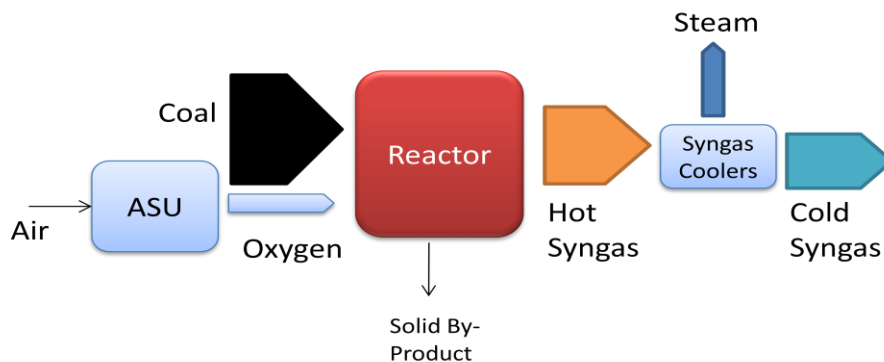


Figure 6. Exergy flows in coal gasifiers

Table 13. Exergy results

<b>Component</b>	<b>KRW</b> (dry feed fluidized bed)	<b>E-Gas</b> (slurry feed entrained flow)	<b>Shell</b> (dry feed entrained flow)	<b>BGL</b> (dry feed fixed bed)
<b>% Exergy Lost</b>				
Drying	1,0	0,0	1,0	0,0
ASU	2,5	2,5	2,1	2,2
Reactor	17,0	19,8	13,8	14,8
Syngas cooling	9,1	2,1	2,3	0,8
<b>Exergy Balance</b>				
Exergy Input, MW	284	882,5	815	358
Exergy from steam production, MW (from syngas coolers)	0	26,2	51,5	5,8
Exergy from syngas production, MW	200	641	607	288,49
<b>Exergy conversion efficiency, %</b>	<b>70,4</b>	<b>75,6</b>	<b>80,8</b>	<b>82,2</b>

#### 2.2.4 Effect of coal type on the exergy conversion efficiency

The exergy conversion efficiency for bituminous and sub bituminous coal is presented in table 14, where it shows that the exergy decreases for lower rank coal. A moisture increase has a more negative impact on the slurry feed than the dry feed entrained flow gasifier because while the carbon concentration in the slurry decrease, the coal can be dried in the dry feeding gasifiers, eliminating partially its effect. While the dry feed gasifier has an exergy efficiency decrease of 2% approximately, the E-Gas slurry has a 9% decrease, mainly because the oxygen amount increases when subbituminous coal is implemented. For the latter reason the coal water slurry gasifiers are recommended only for bituminous coal and for lower rank coal such as sub-bituminous and lignite, the dry feeding systems is recommended in order to maintain high efficiencies. The BGL gasifier wasn't evaluated because the model don't achieve the accuracy to determine the effect of coal type and the KRW operating conditions for subbituminous coal are not available.

Table 14. Exergy conversion efficiency for bituminous and sub-bituminous coal. Sources: [53, 54, 51, 60, 61]

Coal Proximate Analysis		
	Bituminous	Sub bituminous
Moisture, %	11,12	25,77
Ash, %	9,7	8,19
Volatile Matter,%	34,99	30,34
Fixed carbon, %	44,19	35,7
LHV (MJ/kg)	26,15	19,19
Gasifier Inputs		
H <sub>2</sub> O/C (mol)	Shell: 0,11 E-GAS:0,45	Shell:0 E-GAS:0,54
O <sub>2</sub> /C (mol)	Shell:0,41 E-GAS:0,42	Shell:0,44 E-GAS:0,72
Gasifier exergy conversion efficiency		
Shell (dry feed entrained flow)	80,8	78,9
E-GAS (slurry feed entrained flow)	75,6	66,5

### 2.2.5 Capital cost of coal gasifiers

Table 15 shows the most important gasifiers characteristics for the capital cost comparison for bituminous coal gasification. The KWR is not a commercial gasifier because in the Pinon Pine demonstration IGCC plant utilizing this gasifier, it was not able to maintain the syngas production successfully in project period from 1998 to 2000 [64].

The main technology barrier is not technical but economical since the major problems for polygeneration process from coal gasification are the capital cost [66]. In the economic section the cost shown are related for a single gasification train and only the equipment cost are included. Other costs for the plant construction like labor, additional construction materials, taxes or contingencies were not considered. The gasifier section comprises the reactor, the syngas cooler, heat recovery and auxiliaries and the coal preparation and feeding includes the milling, drying, compression and feeding systems.



In order to compare the gasifiers taking into account the capital cost and the coal handling capacity, these two values were divided, founding that the BGL gasifier counts with the lowest value. Although the low coal handling capacity of this technology, for the same capital investment more coal can be gasified with BGL than the other gasifiers. The BGL has lower capital cost because it doesn't dry the coal, has a refractory wall and operates at high temperature only at a section of the reactor where the gasification take place what requires less sophisticated materials in the gasifier [66]. It is the only gasifier with a low waste heat duty with an outlet temperature of 560 °C.

The dry feed entrained flow is the most expensive option but it has to be considered that it is the only technology with membrane water wall which requires less maintenance time and a higher life time [64] [66]. Additionally the cooling screen improves the startup and turndown time and reduces the concerns surrounding coal; especially ash properties due to water membrane wall produce a solid and liquid slag lining instead of a refractory lining [54]. Some coal compound as SiO<sub>2</sub> CaO and iron oxide can penetrate deeply into the high chrome refractory materials and eventually give rise to cracks that lead to material loss [67]. The life of a water wall can provide a significant operating and maintenance cost benefit over refractory lined gasifiers [51].

Table 15. Techno economical comparison of coal gasifiers

<b>Gasification technology</b>	<b>E-Gas</b> (slurry feed entrained flow)	<b>Shell</b> (dry feed entrained flow)	<b>BGL</b> (fixed bed)
<b>Economics USD (\$x1.000)</b>			
Coal preparation and feeding	\$ 11.000	\$ 53.000	\$ 2.500
Air separation unit	\$ 73.000	\$ 74.000	\$ 30.000
Gasifier	\$ 67.000	\$ 93.000	\$ 18.000
Total capital cost	\$ 151.000	\$ 220.000	\$ 50.500
<b>Total capital cost divided by the coal handling capacity (USD \$x1000/Ton-day)</b>	<b>60,4</b>	<b>95,7</b>	<b>50,5</b>

Capital cost Source: [51] [54] [53]

### 3 SIMULATION AND EXERGY COMPARISON OF COAL TO SYNTHETIC NATURAL GAS PRODUCTION PROCESSES

#### 3.1 Processes for the production of synthetic natural gas from coal gasification

The technologies for SNG production can be classified by direct or indirect methanation. The indirect methanation is currently the only available commercial technology and consists in four principal steps; gasification, water gas shift (WGS), gas clean up and methanation. The direct methanation comprises two processes; the hydrogasification and hydromethanation or also called the steam catalytic gasification, which are still considered emerging technologies. The hydromethanation is closed to reach a commercial development and currently a demonstration project at industrial scale is been constructed in China. The hydrogasification is under investigation with a 10 tons per day pilot plant. Table 16 shows the current technology development for the direct and indirect methanation

Currently there are only 2 worldwide SNG plants under commercial operation, one in USA and one in China and both use indirect methanation technology. The plant in China, the Datang, has been recently build and it was confirmed in September 2012 its startup and initial demonstration run [74]. The Great Plains Synfuels Plant in North Dakota USA is the only large scale SNG plant worldwide with significant commercial experience producing methane since 1984.

Table 16. Technology development status of methanation processes (2013)

Technology	Development status	Production plants
<b>Direct Methanation</b>		
Hydromethanation	Large scale development	First demonstration plant is been constructed in China and it is expected to begin operation in 2017
Hydrogasification	Pilot plant investigation	Pilot plant lower of 10 ton per day
<b>Indirect Methanation</b>		
Steam oxygen gasification and downstream methanation	Commercial	Two power plants in operation and numerous under construction

Source: [48, 75] [76, 77] [78, 79] [47]

## **Direct Methanation: Emerging Technologies**

### **Development Status of Hydrogasification Process**

Several test at small pilot plant are under development, investigating the main technical challenges which are the low carbon conversions, low product yields and slow reaction rates [80, 48]. The process was tested in 2006 at a bench level by the Viresco Energy and the company currently works (2011 last report) to build a pilot plant in Utah, a small scale plant with a capacity for 5 tons per day of coal or a mixture coal/biomass to evaluate the steam hydrogasification technical feasibility [48, 75]. Another similar project is been developed by the Arizona Public Service with the financial support of the department of energy with a small pilot plant. Two of the total three project phases are accomplished which tries to determine if the hydrogasification without steam injecting could be a commercially viable process [48]. The successful of these projects could provide the engineering information to develop bigger plants.

### **Development Status of the Hydromethanation Process**

The development status of the hydromethanation technology is under large scale demonstration [78, 79] and in 2017 the first large scale plant in the world is expected to complete its construction in China [76, 77]. The first catalytic gasification process was demonstrated by Exxon in 1979 with a one ton per day pilot plant [79]. The process was improved by the Great Point Energy Group with the hydromethanation, developing more than 60 patents on the technology. The project was demonstrated at near commercial condition with a 100 foot reactor in 2007 and currently conducts test to demonstrate the latest version of their technology to overcome some technical problems [78]. The main challenges to overcome are the catalyst poisoning diminishing its performance over time, the catalysts cost and difficulties recovering and recycling the catalyst [47, 79].

### **Development status of the Indirect methanation: Commercial Technologies**

The commercially proved SNG technology is based on traditional oxygen-steam gasification technologies operated to yield multiple products from syngas. From the total syngas production, 45% is employed to produce chemicals like fertilizers, 38% for liquid fuels, 11% for electric power and only 5% to produce methane [77]. Apparently the actual

SNG commercial process was adapted from the existing technology and wasn't design directly to produce methane.

#### *Methanation catalyst for syngas reforming*

Extensive catalyst metals investigations have been developed mainly on nickel, cobalt, iron, ruthenium, molybdenum and tungsten materials. The preferred catalyst material is the nickel based due to the high activity and low price and its main disadvantage is the extreme vulnerability to sulfur poisoning. A syngas with 20 sulfur ppb gives a nickel life cycle of 4 years approximately [45, 81]. Another important technical challenge is the catalyst temperature resistance. The allowed temperature for nickel commercial catalyst is in the range of 240-650C [45, 50].

## **3.2 Simulation and Exergy Analysis of Coal to SNG Processes**

The process simulated are the indirect methanation and the hydromethanation, both evaluated at chemical equilibrium for all the reactions in the process. The validation is developed only for the indirect methanation since there is no available information for the direct methanation in the open bibliography. The hydrogasification process was not simulated because it is not expected to reach chemical equilibrium and a kinetic model has to be developed which is out of the target of this work.

### **3.2.1 Key Assumptions**

The simulation features described in chapter 3 such as model limitations, scope and coal gasification assumption are also valid for the next simulations. Except the steam and oxygen gasification, all the reactors use catalyst to reach the chemical equilibrium in a short residence time. Therefore the kinetics barriers are decreased and the processes can be evaluated with thermodynamic equilibrium. In the exergetic analysis the oxygen separation degree was in order of 99 percent and all the reactor in the process flow sheets are modeled as adiabatic.

The temperature range of hydromethanation is 600 to 700 C. In this work 600 C since the methane production is favored at lower temperatures [82]. This process uses fluidized bed reactors and technology difficulties as recovering, recycling and poisoning of the catalyst cannot be evaluated with the steady state software. According to Great Point Energy, the

oxygen consumption of hydromethanation is 75 percent lower than the conventional steam oxygen gasification [83] which is equivalent to a maximum value of 0.2 kg oxygen/kg coal bituminous Illinois No. 6. The pressure used in the hydromethanation simulations was 50 bars, the same than the pressure of some steam oxygen gasification plants.

The indirect methanation stages such as the WGS and methanation can use fixed bed reactors since they handle streams without solids. The methanation stage have also difficulties with catalyst poisoning, however in this case the gas is cleaned before with the Selexol process.

Table 17. Key Assumptions

	Direct methanation: Hydromethanation	Indirect Methanation
<b>Gasifier</b>		
Technology	Great Point Energy	Shell
H <sub>2</sub> O/C (mol)	1	0,01
O <sub>2</sub> /C (mol)	0,11	0,44
Oxygen purity	99%	99%
Carbon conversion	Calculated with the model	99%
Temperature	600 C	Calculated with the model
Pressure (Bars)	50	50
<b>Global simulation parameters</b>		
Physical property method	Peng Robinson	Peng Robinson
Kinetics model	Chemical Equilibrium	Chemical Equilibrium
Heat loss	Adiabatic (Except the gasifier)	Adiabatic
Pressure loss (MPa)	ASU: 0,2 Gasifier: 1,3 WGS: 0,45 Methanation: 0,61 Gas clean up: 0,07	ASU: 0,2 Gasifier: 1,3 Gas clean up: 0,07
<b>Indirect methanation parameters</b>		
Equilibrium Temperature 1/2/3 Stage		WGS: 428/252 C
		Methanation: 600/ 438/303 C
Steam Cycle 1/2/3 Stage		Pressure (MPa): 12,5/3,45/0,44 Temperature (C): 565/534/260
<b>Coal: Bituminous Illinois # 6</b>		
Moisture, %	11,12	
Ash, %	9,7	
Volatile Matter,%	34,99	
Fixed carbon, %	44,19	
LHV (MJ/kg)	26,15	

The inputs to the model are summarized in table 17 for the direct and indirect methanation processes. The most important input to the reactors is the equilibrium temperature which is limited by the fixed bed reactors and by the catalyst technology. The gasification technology used for the exergy comparison was the Shell gasifier since it has one of the highest cold gas efficiencies.

## Description of the model flow sheet

### Indirect Methanation: Water gas shift and methanation

The model flow sheet of the water gas shift and the methanation stage are shown in figure 7 and 8 respectively. All the reactors are Gibbs reactors which calculates the equilibrium of the streams for a given temperature and pressure. Initially the syngas produced in the gasifier is mixed with steam in order to have a CO/H<sub>2</sub>O ratio of 1 and send to the water gas shift reactor which consists of two adiabatic Gibbs reactors with inter cooling heat exchanger. In the last heat exchanger most of the water is condensed and recycled to the process. The heat duty from the heat exchanger is used to produce steam to the process. The gas cleanup is not shown on the figures and it is simulated as a simple separator unit which removes the CO<sub>2</sub> and the sulfur components. The methanation flow sheet is similar to the water gas shift but this unit uses three fixed bed reactors to produce high purity methane and use a compressor to recycle some of the gas to control the inlet temperature to the first reactor. The gas is also cooled with steam for power production and the last heat exchanger condenses the water. While the temperature in the gasification model can be calculated from the coal heating value, the other reactors need the equilibrium temperature as a model input.

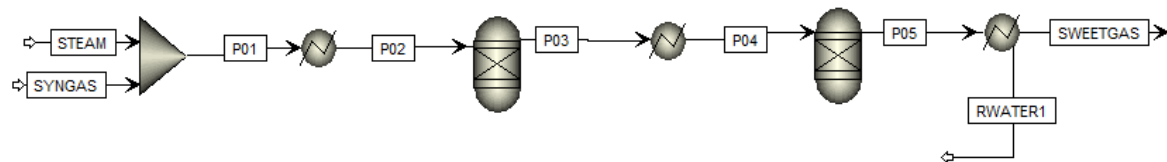


Figure 7. Water gas shift reactors

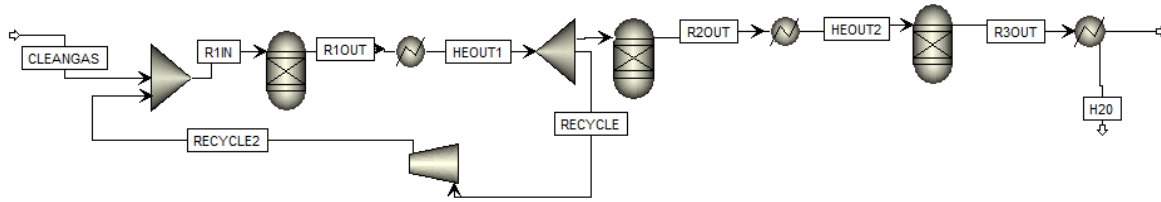


Figure 8. Tremp Methanation Process

## Direct Methanation

The hydromethanation flow sheet is similar to the conventional steam oxygen gasification flow sheet and the only difference is that the flow sheet of figure 9 does not show the dotted line from the yield to the Gibbs reactor because the temperature is a model input not calculated with the software. The hydromethanation uses catalyst to enhance the production of methane and the reaction rates at a temperatures range of 600 to 700 C [82]. The processes reach the chemical equilibrium, therefore can be modeled with a Gibbs reactor and the unreacted char is recycle to ensure complete carbon conversion.

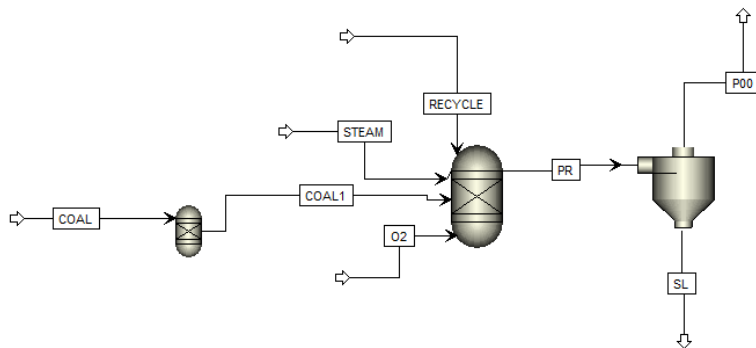


Figure 9. Hydromethanation

### 3.3 Simulation Results

#### Methanation

The simulation results are compared with the reported data from the bibliography as shown in table 18. The model has a high accuracy with an error less than 1 percent for each process stage. The outlet stream of the third stage contains a high steam fraction, which has to be condensed to reach a methane purity more than 85 percent. Additionally the CO<sub>2</sub> can also be separated to purify even more the product stream.

Table 18. Verification of the model

	First stage		Second stage		Third stage	
	Simulation	Reference [84]	Simulation	Reference [84]	Simulation	Reference [84]
H <sub>2</sub> O	31,1234989	31,0	40,2	39,9	43,7593435	43,4
CH <sub>4</sub>	38,0927144	38,1	45,0	44,7	47,4333531	47,2
CO	1,1469081	1,0	0,0	0	0,00108465	0
CO <sub>2</sub>	4,92054739	5,0	2,6	2,4	1,40811985	1,4
H <sub>2</sub>	19,1394027	19,1	6,1	6,7	1,22983486	1,6
N <sub>2</sub>	5,57692853	5,8	6,0	6,3	6,16826409	6,4

#### Hydromethanation

There are no available experimental data in the bibliography for the hydromethanation process since this process is still on experimental state. The simulations were performed at 3 operating pressures and it can be seen that higher pressures benefits the methane production, from 10 to 50 bars it enhances 3 mol percent on the final composition. It is expected that the process reach the chemical equilibrium with the catalyst implemented similarly to the Tremp methanation process. On the other hand the minimal gasification temperature to reach complete carbon conversion is 1000 C. For this reason with the low operating temperature of 600C, 43 percent carbon moles do not react and has to be recycling in the gasifier. The final gas compositions are composed mainly of hydrogen, steam, carbon dioxide and methane as shown in table 19.



Table 19. Equilibrium compositions of the hydromethanation process at 3 different operating pressures

Hydromethanation - Mol %			
	50 bars	30 bars	10 bars
H <sub>2</sub> O	32,7	31,4	28,1
N <sub>2</sub>	0,4	0,4	0,4
H <sub>2</sub>	7,6	9,7	15,5
CO	1,4	1,8	3,1
CO <sub>2</sub>	15,3	15,3	15,1
H <sub>2</sub> S	0,7	0,6	0,6
CH <sub>4</sub>	17,2	16,3	13,6
Carbon/Char	24,7	24,4	23,6

### 3.4 Exergy Performance Results and discussion

The exergy performance results of the direct and indirect methanation are shown in table 20 where it states that 29,3 percentage of the coal exergy is lost in the hydromethanation process and 42,9 percent in the indirect methanation, equivalent to an exergy efficiency of 70,7% and 57,1% respectively. The higher efficiency of hydromethanation responds to the stage reduction, lower exothermic reactions with a more neutral thermal balance of the reactions and lower temperature gap between the gasifier and clean up equipment. The conventional oxygen-steam gasifiers produce syngas that can be upgraded to methane but with a more exergetic expensive product than the direct methanation because of the greater number of stages needed. The sensible heat of the syngas is partially recovered in the indirect methanation with the steam turbine, however the syngas production is more suitable for other products like electricity production [78].

Additionally the technology of hydromethanation converts more coal to methane since while the direct methanation needs 2 carbon moles to produce 1 mole of methane, the indirect methanation needs almost 3 carbon moles to produce 1 mole of methane. The difference between carbon moles are compensated with the carbon dioxide production since more carbon dioxide is produced in the indirect methanation with a CO<sub>2</sub>/C ration of 0,785 and in hydromethanation the ratio is equal to 0,474.

As a result of the higher carbon dioxide production in the indirect methanation a higher fraction of gas has to be separated in the gas cleanup stage, producing 4,3 percent more exergy loss than the direct methanation.

The air separation unit of hydromethanation in table 20 includes the ASU for oxygen separation and methane purification. It has less exergy losses than the indirect methanation ASU mainly because hydromethanation has 75 percent less oxygen consumption than the conventional gasification.

Although the significant difference of the total efficiency between the two processes, it has to be considered that in the hydromethanation process presents some intrinsic thermodynamic limitations such as the carbon conversion goes only up to 57% while for the indirect methanation reach 99%, because the chemical equilibrium constants are higher for the oxygen gasification than for the steam catalytic gasification. The low carbon conversion implies that higher reactor volumes has to be used because more unconverted coal has to be recycle, decreasing the coal handling capacity. Therefore high gasification capacities of 2500 tons/day like conventional coal gasification are not expected to be constructed. In this work the same coal handling capacity was assumed because there is no available information in the bibliography and to compare easier the two plants. Besides the hydromethanation still presents challenging technical difficulties mainly on the catalyst poisoning diminishing its performance over time and recovering and recycling the catalyst which don't take place in the indirect methanation.

Table 20. Exergy balance in coal to SNG processes

Indirect methanation			Direct methanation		
Exergy results					
Process Stage	% Exergy loss	Exergy losses (MW)	Process Stage	Efficiency Defect	Exergy losses (MW)
ASU	2,5	14,4	ASU	1,5	8,6
Gasifier	21,1	121,1	Hydromethanation reactor	24,4	140,1
WGS	3,0	17,2			
Methanation	4,7	27,0			
Gas clean up	7,7	44,2	Gas clean up	3,4	19,5
Steam turbine	3,9	22,4			
Total	42,9	246,2	Total	29,3	168,2
Plant performance					
Exergy input (MW)		574,0	Exergy input (MW)		574,0
Exergy output (MW)		327,8	Exergy output (MW)		405,8
Exergy conversion efficiency (%)		57,1	Exergy conversion efficiency, %		70,7
Methane and Carbon Dioxide Production					
CH4/C ratio (mol)		0,387	CH4/C ratio (mol)		0,505
CO2/C ratio (mol)		0,785	CO2/C ratio (mol)		0,474

### **3.5 Sustainability of the indirect methanation**

#### **3.5.1 Outlook of the next SNG plants**

The economic viability of the coal to SNG projects depends mainly on the natural gas local price, secondly on the coal price and the technology implemented [85]. Several facilities were planned in the United States in the time frame 2005-2006 when natural gas prices were floating between 12-13 dollars per million BTUs, but the prices unexpectedly fell as a result of the enormous unconventional gas (shale gas) resources founding in 2008-2009. In 2012 the natural gas prices reached 2 dollars per million BTUs [86] [87] [88], consequently in USA the coal to SNG plants are been driven out of business with 9 SNG projects canceled or put on hold [89].

On the other hand China has a lack of natural gas resources while the demand has become increasingly prominent [90]. The situation increases the natural gas prices in some Chinese regions up to 12 dollars per million BTUs, around 5 times more expensive than USA market [91]. Since the costs of producing SNG via steam-oxygen commercial coal gasification is estimated to be from 6.7 to 7.5 dollars per million Btus [85], it has a quite economic viability in China and this country may have most of the next SNG plants. In total there are 80 SNG from coal plant projects planning and under construction in China [92] [93]. The first construction phase will produce 198.041 billion cubic meter per year and after the second construction phase the plants will be able to supply 248.896 bcm, 200% of the total China consumption in 2011 (129 bcm) [92].

#### **3.5.2 Capital investment**

The main barrier of the indirect methanation technology is that it requires a high capital investment, high maintenance cost and has a low availability generally in the first years of operation [64]. In order to reach availabilities over 90%, the plant has to count with spare gasifier what makes the plant even more expensive. For these reasons, many gasification projects still need the government economic support [43]. It is expected that the hydromethanation process can reduce the capital and operational costs since it has less stages.

The plant capital costs were reported by the US department with an expected accuracy of -15% to 30% on the low side and a +20 to 50% on the high side [54]. These costs correspond to the total amount needed for the plant construction including the equipment,

labor, additional construction materials, taxes and contingencies, which are shown in table 21. Financial costs and first consumable costs were not considered.

The SNG plant studied operates six of the biggest commercial Shell gasifier of 757 MW coal input, three train Selexol gas clean up technology and three TREMP methanation units to produce approximately 156 million of standard cubic feet SNG.

The most expensive component of SNG plant is the gasifier with the 45,6% capital investment. The gasifier selected presents high cost mainly as a result of the gasifier features like coal drying, pulverizing and dry coal injection system to achieved high efficiency and coal handling capacities. The capital cost of CO<sub>2</sub> removal and compression is low as a result of the precombustion separation system.

Table 21. Capital investment of indirect Methanation

**USD\$ \*1000**

<b>Component</b>	<b>Capital Investment</b>	<b>%</b>
Sorbent handling and feed water	\$ 98.009	3,6
Gasifier and accessories	\$ 1.230.380	45,6
Air separation Unit	\$ 288.039	10,7
Gas Cleanup and piping	\$ 625.812	23,2
CO <sub>2</sub> compression	\$ 71.602	2,7
Methanation and shift reactors	\$ 99.784	3,7
Steam turbine and accessories	\$ 156.800	5,8
Cooling water systems	\$ 45.274	1,7
HRSG, ducting and stack	\$ 5.096	0,2
Instrumentation and Control	\$ 30.286	1,1
Building and Structures	\$ 47.277	1,8
<b>Total cost</b>	<b>\$ 2.698.359</b>	
<b>Production Capacity</b>	<b>156 MPCD</b>	

Source: [54]

### 3.5.3 Operating cost and effect of coal price

The operating and maintenance cost, reported by USDoE in 2011 reached the US\$6.39/MMBtu if the coal have a price of US \$60/ton. The 66% percent of the operating cost corresponds to the fuel and only 34% to other costs like maintenance, operating labor and other consumables. It was assumed that the plant produces its electricity needs and sells the

sulfur produced. Lower rank coals decrease the cost of the SNG from \$6.39/ MMBtu to \$2.85/ MMBtu, when the coal price reduces from \$60/ton to \$10/ton as shown in figure 10.

On the other hand the huge amount of carbon dioxide emissions could impact directly the sustainability of the process if carbon taxes are included in the economy of the country, like European countries which have already included carbon dioxide taxes. In this analysis carbon dioxide taxes were not included.

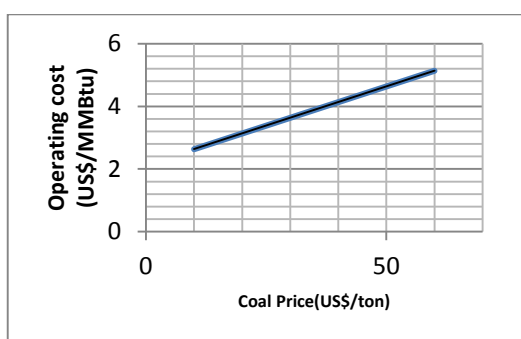


Figure 10. Effect of coal price on the operating cost.

### 3.5.4 Return on investment

The plant capital investment is around \$ 2.698.359.000 US dollars for a production capacity of 155.532 MBtu/day of methane. If the operating cost are \$ 6,39/ MMBtu and the price of natural gas in the international market (Caribbean Region) is \$ 14,0/ MMBtu, the return on investment goes up to 10 years with a capacity factor of 90 percent and 7 percent discount rate, which is a medium period for this large size project. However it has to be considered that the natural gas price can decrease with the revolution of shale gas what risks the project finances.

Table 22 Return on investment

Balance	
Total costs	\$ 2.698.359.000
SNG production capacity	155.532 MBtu/day
Operating cost	\$ 6,39/ MMBtu
Natural gas price	\$ 14,0/ MMBtu
Return on investment*	10 Years

\*Capacity factor of 0,9

## **4 EXERGY ENVIRONMENTAL AND CAPITAL COST COMPARISON OF THE INTEGRATED GASIFICATION COMBINED CYCLE WITH THE PULVERIZED COAL RANKINE CYCLE**

### **4.1 Description of the Pulverized Coal Rankine Cycle**

The property streams of the pulverized coal Rankine cycle are collected from simulations from the US Department of Energy in order to develop the exergy analysis

The subcritical and supercritical pulverized coal Rankine cycle (PCRC) consist of the boiler, gas clean up and the Rankine cycle. The main difference between the two plants is the operating pressure and temperature conditions of 16,5 MPa and 566 °C for the subcritical case and 24.1MPa and 593 °C for the supercritical case. It has to be considered that there is under investigation an ultra supercritical Rankine Cycle with a working temperature of 760 °C and pressure of 34,5 MPa, but this technology is not already available and it is still under investigation [51]. In carbon capture cases an additional Fluor Econamine FG unit is used and consists of a formulation of MEA and proprietary corrosion inhibitor to recover the CO<sub>2</sub> by absorption from the flue gas.

### **4.2 Simulation of the Integrated Gasification Combined Cycle - IGCC**

#### **4.2.1 IGCC key assumption**

Two gasifiers were selected in order to evaluate the IGCC performance, the Shell and E-Gas gasifiers, both entrained flow reactors. Both gasifiers count with heat recovery for steam production in the non capture case and a water quench for the capture case. The dry feed gasifier is used as base case, thus this technology is analyzed for all the section in the study, except in the exergoeconomic analysis where the slurry feed gasifier is also analyzed.

The gasification agents are steam and 95% pure oxygen separated with cryogenic distillation air separation unit (ASU). The nitrogen separated in the ASU, is send to the gas turbine for syngas dilution in order to avoid NO<sub>x</sub> formation.

After the particulate removal the syngas passes through the acid gas cleanup stage, where the Selexol process was analyzed because it has a wider commercial experience. Finally the syngas is burned in an Advance F class gas turbine and with the flue gases, steam is generated which are used to produce electricity in a three pressure stage subcritical steam turbine. The model flow sheet and the key assumptions for the gas turbine and the Rankine cycle are shown in figure 11 and table 23.

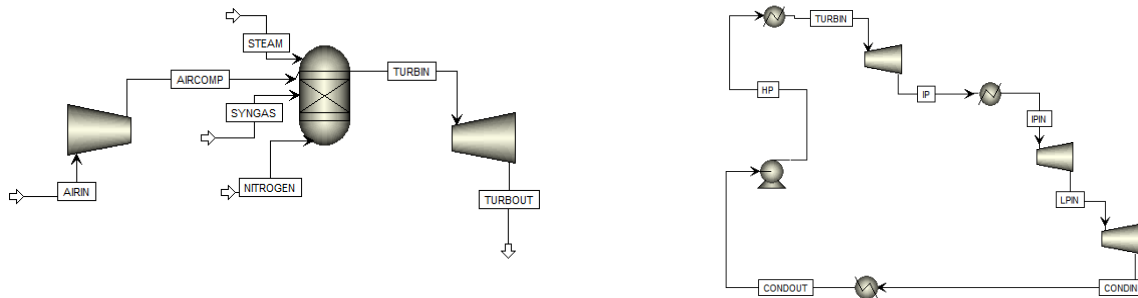


Figure 11. Model flow sheet of the Combined Cycle

Table 23. Key Assumptions

Advanced F Class Combustion Turbine	
Firing Temperature for Syngas (Natural Gas), °C	1338 (1371)
Airflow, kg/s	431
Pressure Ratio	18,5
Net Output, MW	280
Net Efficiency (LHV), %	57,5
Net Heat Rate (LHV), kJ/kWh	6,256
Rankine Cycle	
Steam Turbine 1/2/3 Stage	Pressure (MPa): 12,5/3,1/0,44
	Temperature (C): 558/558/260
Condenser Pressure, kPa	6,8
Pinch Temperature in HRSG, C	30
Pressure Lost in HRSG, Bar	30
Other Stages	
Gasifier Technology	Shell/E-Gas
ASU	95 % Oxygen
Gas Clean Up	Selexol

### 4.3 Exergy results and comparison between IGCC with and without CO<sub>2</sub> capture

Table 24 shows the exergy balance for the proposed IGCC plant with and without CO<sub>2</sub> capture. The capture case has 9.5 % higher exergy losses than the non capture case mainly because the higher entropy production in the cleanup section and the water gas shift reactors. The shift reactors are included only for the capture case to convert the carbon monoxide into hydrogen, avoiding the production of CO<sub>2</sub> in the combustion chamber. The CO<sub>2</sub> produced in the water gas shift reactor corresponds to the 37% of the syngas mole fraction which accounts as a significant amount of exergy destroyed when is separated in the gas cleanup section. Besides there is approximately 60% more energy consumption in the process mainly from the additional CO<sub>2</sub> compression and a significant increased in the acid gas removal. Consequently the fraction of the exergy lost in the clean up section and water gas shift reactors increase 5,6 and 1,9 % respectively.

The difference between the two cases in the gasifier and the air separation unit are 1,8 % and 0,8% respectively higher in the CO<sub>2</sub> capture case due to the gasifier cooling section in this case uses a water quench to have a high H<sub>2</sub>O/CO ratio for the downstream shift reaction. On the other hand in the non-capture case the syngas high sensible heat is partially recovered for steam generation and send to the HRSG. In addition the integration between the air separation unit and the turbine compressor is recommended to enhance the global efficiency and 25 to 30 percent of the ASU air from the turbine provides the best balance between plant output, availability, efficiency and reliability [51, 52]. However for hydrogen syngas corresponding to CO<sub>2</sub> capture case, all of the available combustion air is required in the gas turbine to maintain a high performance.

The gas turbine also have a higher entropy production for the capture case because of the turbine firing temperature has to be reduced to allow parts having an equivalent lifetime [51].

Thermodynamically with a higher turbine inlet temperature, higher will be the efficiency, however the turbine firing temperature is limited to metallurgy restriction.

Syngas with high hydrogen content produced water in the combustion which increases the heat transfer to the turbine blades due to the higher heat transfer coefficient of water, thus the firing temperature has to decrease to maintain the same turbine lifetime [64]. For the same turbine lifetime the firing temperature has to decrease 20 C when the hydrogen fraction increased from 0.31 to 0.985 what produces in combustor exhaust gases a H<sub>2</sub>O molar fraction increased from 0.0719 to 0.1307. As a results of this the difference of exergy lost between the two cases is 0,8 %.



The only components with a lower Irreversibility production in the capture case are the heat recovery steam generation (HRSG) and the steam turbine, because in this case no steam is produced in the gasifier and there is less steam sources to mix, producing less irreversibilities.

The components with the highest electricity consumption are the air separation unit with 32%, the CO<sub>2</sub> compressor with 18 % and the coal feeding system with 17 %. However the exergy lost for the air separation unit is the one of the lowest because the biggest plant irreversibilities corresponds to the components with the combustion reaction in the gasifier and the gas turbine combustion chamber. The gasifier has less entropy production than the gas turbine because the gasification is an incomplete combustion of the coal and in the turbine the fuel is complete oxidized.

Table 24. Exergy lost in IGCC plants with and without CO<sub>2</sub> capture

<b>IGCC (Dry Feed entrained flow gasifier)</b>		
	without CO <sub>2</sub> capture	with CO <sub>2</sub> capture
Exergy conversion efficiency, %	40,1	30,6
<b>Process stage</b>	<b>% Exergy lost</b>	
Gasifier	19,3	21,1
ASU	1,7	2,5
WGS	N/A	1,9
Gas clean up	4	9,6
Gas turbine	22,1	22,9
HRSG and Steam turbine	9	7,9
Stack	3,8	3,5
<b>Total</b>	<b>59,9</b>	<b>69,4</b>

## **4.4 Comparison between IGCC, subcritical and supercritical pulverized coal rankine cycle with and without CO<sub>2</sub> capture**

### **4.4.1 Exergoeconomic analysis**

The exergy conversion efficiency and capital cost of the plant vary significantly with different coal types and if CO<sub>2</sub> capture and sequestration is implemented. For all cases the IGCC plant has a slight higher exergy conversion efficiency of 1 to 5% than the pulverized coal Rankine Cycle (PCRC) plants and given that the gasification plant has more stages, it is more expensive but only for the CO<sub>2</sub> non capture case, as shown in table 25. The capital cost are at least 16 % higher for IGCC when the plant does not use carbon dioxide capture but it has a lower cost with the slurry feed gasifier when the CO<sub>2</sub> emissions need to be sequestered. The IGCC process depends strongly on the gasifier technology and it can be seen that the slurry technology has a lower capital requirement with CO<sub>2</sub> capture than the PCRC. The dry feed gasifier is always more expensive but has the highest exergy efficiency. The gasification plant has a better economical and exergetical performance for CO<sub>2</sub> capture cases mainly because of the pre combustion gas clean up separates the undesired gases at a lower temperature and higher pressure with an elevated density. The conventional pulverized coal plants has to handle approximately 10 times more gas in the post combustion clean up stage. The cost of implementing CO<sub>2</sub> capture in pulverized coal plants is approximately 1300 \$/kW and in IGCC 900 \$/kW, for a total difference of 400 \$/kW more expensive. However the operating costs are for all cases more expensive for the IGCCC process because it presents more process units and stages.

The dry feed gasifier has a greater negative impact on the exergy efficiency when the greenhouse emissions are capture, because of its dry feeding system produce a syngas composition with a lower H<sub>2</sub>/CO ration than the slurry gasifier. The syngas composition is relative insensitive of coal type and is mainly a function of the gasification technology, the reactor type and the operating conditions [45, 64]. This ratio should be as high as possible in order to have a better performance in the downstream water gas shift reactors. In case that the syngas has low hydrogen percentage, the carbon dioxide production in the WGS increases what reduces the global plant efficiency.

Additionally in carbon capture application, the dry feed gasifier compressed the coal with nitrogen and with higher nitrogen content, the raw gas is more diluted and therefore reduces the CO<sub>2</sub> partial pressure [64]. For these reasons the slurry gasifiers is recommended when carbon capture is applied.

The PCRC cannot process low rank coals since the emissions can exceed the environmental requirements. On the other hand a big advantage of the IGCC process is the capacity of

handling low rank coals with high environmental performance. The exergy efficiency decreases for lower rank coals mainly because the gasifier cold gas efficiency decreases with higher ash and moisture content.

A moisture or ash increase has a more negative impact on the slurry feeding system than the dry system because while the carbon concentration in the slurry decrease, the coal has to be dried in the dry system [64], eliminating partially its effect. Comparing the two coal cases without CO<sub>2</sub> capture, the IGCC with the dry feed gasifier has a efficiency decrease of 0,8% while the slurry case has a 3,6% decrease. The dry feed gasifier presents an exergy advantage for lower rank coal with no carbon capture and this difference is expected to be higher for lignite. Thus to maintain the highest efficiencies, the dry feed gasifier are recommended for lower rank coals when no carbon capture is implemented.

Additionally for lower rank coals the cost increases since for the same production capacity more coal has to be processed what require larger equipment size to handle syngas with lower density. Considering the IGCC plant, the cost increase to change from bituminous to sub bituminous coal is approximately 300 \$/kW for all cases.

Table 25. Comparison between IGCC, subcritical and supercritical pulverized coal rankine cycle with and without CO<sub>2</sub> capture

	IGCC		Subcritical	Supercritical
	Shell	E-GAS		
Bituminous Coal				
Capital costs				
Without CO2 capture, \$/kW	2.217	1.913	1.622	1.647
With CO2 capture and sequestration, \$/kW	3.181	2.817	2.942	2.913
Operating Costs				
Without CO2 capture, \$/kW (net)	85	77	59	58
With CO2 capture and sequestration, \$/kW	117	108	96	97
Exergy conversion efficiency				
Without CO2 capture, %	40,1	38,0	35,4	37,5
With CO2 capture and sequestration, %	30,6	29,7	25,2	27,4
Subbituminous Coal				
Capital costs				
Without CO2 capture, \$/kW	2.506	2.265	N/A	
With CO2 capture and sequestration, \$/kW	3.480	3.144		
Exergy conversion efficiency				
Without CO2 capture, %	39,3	34,4	N/A	
With CO2 capture and sequestration, %	30,0	28,5		

Economic Source: [51, 53, 54]

#### 4.4.2 Exergy comparison of IGCC and pulverized coal plants at component level

The exergetic performance of the principal process components for the IGCC, the subcritical and supercritical pulverized coal Rankine cycle plant is shown on the table 26. The dry feed gasifier was selected for the IGCC plant since it has the highest exergy efficiency although it presents the highest costs. For the three plants the main irreversibilities source corresponds to the combustion reactions in the gas turbine, the gasifier and the boiler.

As shown in the latter section the IGCC process presents less irreversibilities than the other two plants. In spite of the big irreversibility contribution of the gasifier, the IGCC is more exergy efficient due to the combine cycle have a better exergy performance than the rankine cycle and more exergy is lost in the stack gases for the pulverized coal plant. The irreversibilities of the IGCC non-capture case are reduced up to 4,7 % compared with the subcritical plant and 2,2 % with the supercritical process. Regarding the carbon capture cases, the differences between IGCC and subcritical are 5,4 % and 3,2 % compared with the supercritical case. The difference is specifically advantageous for IGCC when CO<sub>2</sub> capture is applied due to the exergy difference is significant higher.

The IGCC gas clean up has a higher Irreversibilities than the other plants mainly as a results of the separation of the H<sub>2</sub>S with a higher exergy content than SO<sub>2</sub> and additionally the CO<sub>2</sub> separation before the gas turbine which does not allow the work production from these gases.

Table 26. Percentage of exergy lost of IGCC and pulverized coal Rankine cycle plants at component level

Process stage	IGCC (Dry Feed Gasifier)		Process stage	Subcritical PC rankine cycle		Subcritical PC rankine cycle	
	With CO2 capture	Without CO2 capture		With CO2 capture	Without CO2 capture	With CO2 capture	Without CO2 capture
Gasifier	21,1	19,3	Boiler	48,6	48,6	47	40,8
ASU	2,5	1,7					
WGS	1,9	N/A					
Gas clean up	9,6	4	Gas clean up	4,1	0,7	4,1	0,7
Gas turbine	22,9	22,1	Steam turbine	16,9	10	16,4	15,4
HRSG and Steam turbine	7,9	9					
Stack	3,5	3,8	Stack gases	5,2	5,2	5,2	5,2
Total	69,4	59,9	Total	74,8	64,6	72,6	62,1

#### 4.4.3 Environmental Performance

The gas emission are illustrated in table 27, where it shows that for all plants the emissions of SO<sub>x</sub>, NO<sub>x</sub>, Hg and particulates decrease implementing the carbon capture and IGCC have a better environmental performance, specially for the non capture case. The sulfur dioxide and carbon dioxide emissions are almost the same in the three plants for the capture cases. The NO<sub>x</sub> production is significant lower for IGCC due to this emission can be controlled with nitrogen dilution, humidification and steam dilution [51].

While the pulverized coal plants have to use a postcombustion gas clean up stage, the IGCC plant can use precombustion carbon capture, but has to separate the sulfur species and particulates before the combustion due to the turbine lifetime and operational safety [94] [95]. The precombustion option is more energy efficient because it has a lower net energy consumption as a result that the gas is cleaned at a higher density when it is still compressed and at a cold temperature. The electricity consumption, illustrated in table 27, includes the acid gas removal, the scrubber pumps, the Claus plant and the CO<sub>2</sub> compressor in capture cases for the IGCC plant and the induced fans, Wet Limestone Forced Oxidation and CO<sub>2</sub> compressor for the pulverized coal rankine cycle plants. The electricity consumption is always higher for the pulverized coal plants, reaching 30 percent higher electricity consumption in carbon capture applications and go up to 4 to 5 times higher in non capture cases.

The table 27 comprises the emissions for bituminous coals, since for lower rank coals cases the pulverized coal plants increases the emissions considerably and therefore these plants are recommended only for bituminous coals.

For the IGCC cases the gas emissions maintain practically constant for lower rank coals including subbituminous coals and lignite [51, 53]. The only significant emission increased is presented for the CO<sub>2</sub> component for the non-capture cases where the amount rise from 617 to 647 kg/MWhe, what represents 5% more greenhouse emissions mainly due to the higher irreversibility produced in the gasifier oxidizing the coal.

Table 27. Environmental performance of IGCC and pulverized coal rankine cycle plants

		IGCC		Subcritical PC rankine cycle		Subcritical PC rankine cycle	
		With CO2 capture (90%)	Without CO2 capture	With CO2 capture	Without CO2 capture	With CO2 capture	Without CO2 capture
SO <sub>2</sub>	[kg/MWhe]	0,008	0,013	0,008	0,341	0,007	0,32
NO <sub>x</sub>		0,18	0,185	0,339	0,278	0,316	0,261
Particulates		0,026	0,022	0,063	0,052	0,059	0,049
Hg		2,09E-06	1,79E-06	5,53E-06	4,54E-06	5,16E-06	4,27E-06
CO <sub>2</sub>		73	617	98	809	92	760
Gas clean up electricity consumption	kWh/kg coal	243	13	319	55	319	59

Source: [51, 53, 54]

## 5 Conclusions

The main conclusions of the study are classified in three components by methane, electricity and syngas production in order to respond the following investigation problem;

**How much can the emerging technologies increase the exergy performance, the sustainability and decrease the greenhouse emissions of power and fuels production derived from coal?**

### Methane production

The hydromethanation process has an exergetic conversion efficiency of 70,7%, and the conventional indirect methanation technology goes up to 57,1%, for a total 13 % difference. In addition the indirect methanation has a higher carbon dioxide production of 0,785 CO<sub>2</sub>/C mole ratio compared to the 0,47 CO<sub>2</sub>/C mole ratio of the direct methanation.

The current commercial indirect methanation technology is a risky option considering that is a high investment technology with a high carbon dioxide production, extremely vulnerable to natural gas local price. The hydromethanation technology is expected to demonstrate its feasibility by 2017 and can eventually lower the process capital costs.

### Electricity production

The exergy performance of IGCC with the dry feed gasifier is 2,6 % higher than the supercritical PCRC for the CO<sub>2</sub> non capture case and 3,2 % higher for the capture case. The sources of the biggest irreversibilities correspond to the combustion reactions in the gas turbine, the gasifier and the boiler.

The IGCC plant can be more economical than the PCRC when CO<sub>2</sub> capture and sequestration is implemented, depending which gasifier technology is used. However the operating costs are for all cases more expensive for the IGCC process because it presents more process units. The IGCC has total greenhouse gas emissions of 617 kg CO<sub>2</sub>/MWhe, 143 lower than the supercritical pulverized coal Rankine Cycle which goes up to 760 kg CO<sub>2</sub>/MWhe. The greenhouse gas emissions is lower for IGCC mainly due to the higher exergy conversion efficiency of the coal gasification plant and the precombustion CO<sub>2</sub> capture system.

The IGCC process can be slightly more advantageous depending on the gasification technology and only when CO<sub>2</sub> capture is implemented.

## **Syngas production**

The coal gasifiers has an exergy conversion efficiency in the range of 72 to 82 % for bituminous coals and presents the highest capital cost fraction in the plant with 36 and 46% of an IGCC and indirect methanation plant respectively. The exergy conversion efficiency decreases 3% for the dry feed gasifier and 9 % for the slurry gasifier when instead of bituminous, subbituminous coal is implemented.

Considering the technical, economical aspects of coal gasifiers, the fixed bed gasifier represents the most suitable option, comprising the highest exergy conversion efficiency of 82 % and although it has the lowest coal handling capacity, the fixed bed gasifier produces more syngas than the entrained flow gasifiers with the same capital investment.

Considering the environmental performance, the slurry gasifier has the highest CO<sub>2</sub> emissions production and the fixed bed reactor has the highest pollutant formation.

## *Coal gasification simulation accuracy*

The coal gasification technology was modeled with Aspen plus thermodynamic equilibrium models and the results were compared to the real plants from demonstration projects, founding a medium to high accuracy depending on the gasifier technology. The gasifiers with the highest accuracy are the dry feed entrained flow and the fluidized bed gasifier with less than 2 molar fraction deviation for each chemical component. The slurry feed presents a satisfactory accuracy since the model has up to 3,4 molar fraction deviation and the fixed bed gasifier has the worst accuracy with a 10 percent deviation by the carbon monoxide component. This significant deviation is due to the fixed bed reactors produce hydrocarbons liquids such as tars and oils which are not considered by the model.



## **6 Recommendations for future works**

Natural gas is the most environmental friendly fossil fuel with a high energy content and represent the fuel of the near future. Additionally the direct methanation is currently a emerging technology which could have a great potential converting the most abundant, stable, and low price fossil fuel to a synthetic natural gas. Therefore the hydrogasification process and the hydromethanation are recommended areas for future investigations:

- The hydrogasification kinetics has to be considered in order to evaluate the process. Thus the model construction, validation with experimental data and its evaluation could provide an important contribution to the investigation field. In addition the investigation should focus on how to increase the low carbon conversions, low product yields and slow reaction rates.

- For the hydromethanation process the investigation should focus on the catalyst development such as how to decrease the catalyst poisoning diminishing its performance over time, the catalysts cost due to the rare metals and difficulties recovering and recycling the catalyst in the reactor.

## 7 References

- [1] T. J. Kotas, The exergy method of thermal plant analysis, London: Department of Mechanical Engineering, Queen Mary and Westfield College, University of London, 1985.
- [2] J. Zhang, Z. Zhou, L. Ma, Z. Ni and W. Li, Efficiency of wet feed IGCC (integrated gasification combined cycle) systems with coalwater slurry preheating vaporization technology, Beijing: State Key Laboratory of Power Systems, Department of Thermal Engineering, Tsinghua-BP Clean Energy Center, Tsinghua University, 2012.
- [3] W. Han, H. Jin and R. Lin, A novel power generation system based on moderate conversion of chemical energy of coal and natural gas, Beijing: Institute of Engineering Thermophysics, Chinese Academy of Sciences, 2009.
- [4] M. Kawabata, O. Kurata, N. Iki, A. Tsutsumi and H. Furutani, SYSTEM MODELING OF EXERGY RECUPERATED IGCC SYSTEM WITH PRE- AND POST-COMBUSTION CO<sub>2</sub> CAPTURE, Tsukuba, Ibaraki, Japan: Energy Technology Research Institute, National Institute of Advanced Industrial Science and Technology, 2013.
- [5] De and Biswal, Thermodynamic analysis of a coal gasification and split Rankine combined cogeneration plant. Part 2, exergy analysis, Calcutta, India: Mechanical Engineering Department, Jadavpur University, 2004.
- [6] Zedtwitz and Steinfeld, The solar thermal gasification of coal energy conversion efficiency and CO<sub>2</sub> mitigation potential, Switzerland: Department of Mechanical and Process Engineering, ETH-Swiss Federal Institute of Technology; Paul Scherrer Institute, 2001.
- [7] A. O. Ong'iro, Techno-economic modeling of integrated advanced power cycles, Thechnical University of Nova Scotia, 1996.
- [8] B. Erlach, M. Schmidt and G. Tsatsaronis, Comparison of carbon capture IGCC with pre-combustion decarbonisation and with chemical looping combustion, Berlin: Technische Universität Berlin, Institute for Energy Engineering, 2010.
- [9] G. Zhang, Y. Yang, H. J. b, G. Xu and K. Zhang, Proposed combined-cycle power system based on oxygen-blown coal partial gasification, Beijing: North China Electricity Power University and Institute of Engineering Thermophysics, 2012.
- [10] C. Kunze, K. Riedl and H. Spliethoff, Structured exergy analysis of an integrated gasification combined cycle (IGCC) plant with carbon capture, München: Institute for Energy Systems, 2010.

- [11] N. V. Gnanapragasam, B. V. Reddy and M. A. Rosen, Hydrogen production from coal gasification for effective downstream CO<sub>2</sub> capture, Ontario, Canada: Faculty of Engineering and Applied Science, University of Ontario Institute of Technology, 2009.
- [12] A. Odukoya, I. Dincer and G. F. Naterer, Exergy analysis of a gasification based combined cycle with solid oxide fuel cells for cogeneration, Ontario: Faculty of Engineering and Applied Science, University of Ontario, 2011.
- [13] K. V. LOBACHYOV and H. J. RICHTER, High efficiency coal fired power plant of the future, Hanover: Thayer School of Engineering, Dartmouth College, 1996.
- [14] I. D. G. F. N. Rami Salah El-Emam, Energy and exergy analyses of an integrated SOFC and coal gasification system, Faculty of Engineering and Applied Science, University of Ontario Institute of Technology, Canada, Faculty of Engineering, Mansoura University, Egypt, 2011.
- [15] P. Zedtwitz, J. Petrasch, D. Trommer and A. Steinfeld, Hydrogen production via the solar thermal decarbonization of fossil fuels, Switzerland: Department of Mechanical and Process Engineering, ETH—Swiss Federal Institute of Technology Zurich; Solar Process Technology, Paul Scherrer Institute, 2004.
- [16] Y. Zhu, S. Somasundaram and J. W. Kemp, Energy and Exergy Analysis of Gasifier-Based Coal-to-Fuel Systems, Washington, USA: Energy and Environment Directorate, Energy and Environment Directorate, 2010.
- [17] M. Liszka, T. Malik and G. Manfrida, Energy and exergy analysis of hydrogen-oriented coal gasification with CO<sub>2</sub> capture, Institute of Thermal Technology, Silesian University of Technology; Department of Energy Engineering, University of Florence, 2012.
- [18] Y. Xu, G. Zang, H. Chen, B. Dou and C. Tan, Co-production system of hydrogen and electricity based on coal partial gasification with CO<sub>2</sub> capture, China: Institute of Engineering Thermophysics, Chinese Academy of Sciences, Graduate University of Chinese Academy of Sciences, School of Energy and Power Engineering, Dalian University of Technology, 2012.
- [19] V. I. Anikeev, A. V. Gudkov and A. Yermakova, Theoretical analysis of schemes for complex coal processing, Novosibirsk, Russian Federation: Boreskoc Institute of Catalysis, Akademika Lauretha, 1997.
- [20] P. Gangadharan, A. Zanwar, K. Zheng, J. Gossage and H. H. Lou, Sustainability assessment of polygeneration processes based on syngas derived from coal and natural gas, Beaumont, United States: Dan F. Smith Dept. of Chemical Engineering, Lamar University, 2011.

- [21] Cocco, Pettinau and Cau, Energy and economic assessment of IGCC power plants integrated with DME synthesis processes, Cagliari, Italy: Department of Mechanical Engineering, University of Cagliari, 2005.
- [22] L. Gao, H. Jin, Z. Liu and D. Zheng, Exergy analysis of coal-based polygeneration system for power and chemical production, Beijing, China: Institute of Engineering Thermophysics, Chinese Academy of Sciences; Chemical Engineering College, Beijing University of Chemical Technology, 2004.
- [23] L. Guang-jian, L. Zheng, W. Ming-hua and N. Wei-dou, Energy savings by co-production: A methanol/electricity case study, Beijing, China: School of Energy and Power Engineering, North China Electric Power University; State Key Lab of Power Systems, Dept. of Thermal Engineering, Tsinghua University, 2009.
- [24] D. Yuanyuan, Z. Jin, Z. Mingshan and H. Lizhong, Exergy Analysis of Methanol-IGCC Polygeneration Technology Based on Coal Gasification, Beijing, China: Department of Thermal Engineering, Tsinghua University, 2002.
- [25] S. Sun, H. Jin, L. Gao and W. Han, Study on a multifunctional energy system producing coking heat, methanol and electricity, Beijing, China: Institute of Engineering Thermophysics, Chinese Academy of Sciences; Graduate University of Chinese Academy of Sciences, 2008.
- [26] Z. Li, P. Liua, F. Hea, M. Wanga and E. Pistikopoulosb, Simulation and exergoeconomic analysis of a dual-gas sourced polygeneration process with integrated methanol/DME/DMC catalytic synthesis, State Key Laboratory of Power Systems, Department of Thermal Engineering, Tsinghua University, Beijing, China; Centre for Process Systems Engineering (CPSE), Department of Chemical Engineering, Imperial College London, London, UK, 2010.
- [27] W. Han and H. Jin, A New Kind of Multifunctional Energy System Based on Moderate Conversion of Chemical Energy of Fossil Fuels, Beijing, China: Institute of Engineering Thermophysics, Chinese Academy of Sciences, 2010.
- [28] C. Kunze and H. Spliethoff, Modelling, comparison and operation experiences of entrained flow gasifier, München, Germany: Institute for Energy Systems, TU, 2010.
- [29] I. Janajreh, S. S. Raza and A. S. Valmundsson, Plasma gasification process: Modeling, simulation and comparison with conventional air gasification, Abu Dhabi, United Arab Emirates: Masdar Institute of Science and Technology, 2012.
- [30] G. Zanga, Y. Xua, H. Chena, B. Douc, C. Tana and Z. Lid, Coal Partial Gasification Studies Applied to Co-production of Hydrogen and Electricity, China: School of Energy and Power

Engineering, Dalian University of Technology, National Power Plant Combustion Engineering Technology Research Center, 2012.

- [31] M. Öztürk, N. Özek and Y. E. Yüksel, Gasification of various types of tertiary coals: A sustainability approach, Isparta, Turkey: Department of Physics, Faculty of Art and Sciences, Suleyman Demirel University, 2011.
- [32] S. Bargigli, M. Raugeri and S. Ulgiati, Comparison of thermodynamic and environmental indexes of natural gas, syngas and hydrogen production processes, Siena, Italy: Department of Chemistry, University of Siena, 2004.
- [33] M. J. Prins, K. J. Ptasinski and F. J. Janssen, From coal to biomass gasification: Comparison of thermodynamic efficiency, Eindhoven, The Netherlands: Department of Chemical Engineering, Eindhoven University of Technology, 2004.
- [34] R. J. Dudgeon, An exergy-based analysis of gasification and oxyburn processes, Iowa, USA: Master's thesis, University of Iowa, 2009.
- [35] M. ANHEDEN and G. SVEDBERG, EXERGY ANALYSIS OF CHEMICAL-LOOPING COMBUSTION SYSTEMS, Stockholm, Sweden: Royal Institute of Technology, Chemical Engineering and Technology/Energy Processes, 1998.
- [36] L. v. d. Hama and S. Kjelstrup, Exergy analysis of two cryogenic air separation processes, Norway, Netherlands: Department of Chemistry, Norwegian University of Science and Technology; Department of Process and Energy, Delft University of Technology, 2010.
- [37] R. L. Cornelissen, Thermodynamics and sustainable development The use of exergy analysis and the reduction of irreversibility, Ph.D. thesis, Universiteit Twente, 1997.
- [38] A. Kishimoto, Y. Kansha, C. Fushimi and A. Tsutsumi, Exergy recuperative CO<sub>2</sub> gas separation in pre-combustion capture, Japan: Institute of Industrial Science, University of Tokyo, 2011.
- [39] D. Iribarren, F. Petrakopoulou and J. Dufour, Environmental and thermodynamic evaluation of CO<sub>2</sub> capture, transport and storage with and without enhanced resource recovery, Móstoles, Spain: Systems Analysis Unit, Instituto IMDEA Energía; Department of Chemical and Energy Technology, ESCET, Rey Juan Carlos University, 2012.
- [40] Y. Lara, A. Martínez, P. Lisbona, I. Bolea, A. González and L. M. Romeo, Using the Second Law of Thermodynamic to improve CO<sub>2</sub> capture systems, Zaragoza, Spain: Centre of Research for Energy Resources and Consumption (CIRCE), Thermal Division, 2011.
- [41] L. Lombardi, Life cycle assessment comparison of technical solutions for CO<sub>2</sub> emissions

reduction in power generation, Firenze, Italy: Dipartimento di Energetica 'Sergio Stecco', Università degli Studi di Firenze, 2001.

- [42] USDoE and J. B. Tennant, Overview of DOE's Gasification Program, Available 08.2012 <http://www.netl.doe.gov/technologies/coalpower/gasification/pubs/pdf/DOE%20Gasification%20Program%20Overview.pdf>, 2010.
- [43] J. Phillips, Different Types of Gasifiers and Their Integration with Gas Turbines, Charlotte: Available 08.2012 <http://www.netl.doe.gov/technologies/coalpower/turbines/refshelf/handbook/1.2.1.pdf>, 2004.
- [44] W. H. X. Z. Zhao Yuehong, Conceptual design and simulation study of a co-gasification technology, Beijing, China: Institute of Process Engineering, Chinese Academy of Sciences, 2005.
- [45] J. Kopyscinski, T. Schildhauer and S. Biollaz, Production of synthetic natural gas (SNG) from coal and dry biomass – A technology review from 1950 to 2009, Villigen: General Energy Research Department, Paul Scherrer Institut, 2009.
- [46] S. Lee, Gasification of Coal, Handbook of alternative fuel technologies, Taylor & Francis Group, LLC, 2007.
- [47] M. Chandel and E. Williams, Synthetic Natural Gas (SNG): Technology, Environmental Implications, and Economics, Available 10.2012 from [http://www.nicholas.duke.edu/ccpp/ccpp\\_pdfs/synthetic.gas.pdf](http://www.nicholas.duke.edu/ccpp/ccpp_pdfs/synthetic.gas.pdf), 2009.
- [48] USDoE, Hydrogasification facts, USA: Available 01.11.2012 [http://www.netl.doe.gov/technologies/coalpower/gasification/gasifipedia/4-gasifiers/4-1-4-3\\_hydro.html](http://www.netl.doe.gov/technologies/coalpower/gasification/gasifipedia/4-gasifiers/4-1-4-3_hydro.html), 2012.
- [49] USDoE, Methanation, USA: Available 02.10.2012 [http://www.netl.doe.gov/technologies/coalpower/gasification/gasifipedia/5-support/5-12\\_methanation.html](http://www.netl.doe.gov/technologies/coalpower/gasification/gasifipedia/5-support/5-12_methanation.html), 2012.
- [50] C. Z. L. B. M. F. y. A. B. M. Sudiro, Synthetic Natural Gas (SNG) from petcoke: model development and simulation, Corsico, Milano: Available 11.2012 from <http://www.aidic.it/acos/09/09/036.pdf>.
- [51] USDoE, Cost and Performance Baseline for Fossil Energy Plants Volume 1, Bituminous Coal and Natural Gas to Electricity, Available 08.2012 from [http://www.netl.doe.gov/energy-analyses/pubs/BitBase\\_FinRep\\_Rev2.pdf](http://www.netl.doe.gov/energy-analyses/pubs/BitBase_FinRep_Rev2.pdf), 2010.

- [52] R. F. Geosits and L. A. Schmoe, IGCC - THE CHALLENGES OF INTEGRATION, Houston, USA: Available 11.2012 from <http://www.bechtel.com/assets/files/PDF/BIP/35478.pdf>, 2005.
- [53] USDoE, Cost and Performance Baseline for Fossil Energy Plants Volume 3a: Low Rank Coal to Electricity: IGCC Cases, Available 08.2012 [http://www.netl.doe.gov/energy-analyses/pubs/LR\\_IGCC\\_FR\\_20110511.pdf](http://www.netl.doe.gov/energy-analyses/pubs/LR_IGCC_FR_20110511.pdf), 2011.
- [54] USDoE, Cost and Performance Baseline for Fossil Energy Plants Volume 2, Coal to Synthetic Natural Gas and Ammonia, Available 08.2012 [http://www.netl.doe.gov/energy-analyses/pubs/SNGAmmonia\\_FR\\_20110706.pdf](http://www.netl.doe.gov/energy-analyses/pubs/SNGAmmonia_FR_20110706.pdf), 2011.
- [55] Gasification Technologies Council, Gasification Facts, Extracted on february 19 2013 [http://www.gasification.org/page\\_1.asp?a=85](http://www.gasification.org/page_1.asp?a=85), 2013.
- [56] USDoE, Worldwide Gasification Database, Available 07.2012 [http://www.netl.doe.gov/technologies/coalpower/gasification/worlddatabase/2010\\_Worldwide\\_Gasification\\_Database.pdf](http://www.netl.doe.gov/technologies/coalpower/gasification/worlddatabase/2010_Worldwide_Gasification_Database.pdf), 2010.
- [57] H. C. Mantripragadaa and E. S. Rubina, CO<sub>2</sub> reduction potential of coal-to-liquids (CTL) process: Effect of gasification technology, Elsevier, 2011.
- [58] M. v. d. B. Christopher Higman, Gasification Processes, Second Edition, 2008.
- [59] M. J. Bockelie, M. K. Denison, Z. Chen, C. L. Senior and A. F. Sarofim, Using Models To Select Operating Conditions for Gasifiers, Available 10.2012 [http://www.reaction-eng.com/downloads/REI\\_processmodel.pdf](http://www.reaction-eng.com/downloads/REI_processmodel.pdf).
- [60] USDoE, Industrial Size Gasification for Syngas, Substitute Natural Gas and Power Production, USA: National Energy Technology Laboratory, 2007.
- [61] USDoE, KRW Gasifier IGCC Base Cases, Available 19.09.2013 from [http://www.netl.doe.gov/technologies/coalpower/gasification/pubs/pdf/system/krw3x\\_20.pdf](http://www.netl.doe.gov/technologies/coalpower/gasification/pubs/pdf/system/krw3x_20.pdf), 2000.
- [62] E. C. Carlson, Don't Gamble With Physical Properties For Simulations, Available 07.2012 from [http://www.eq.uc.pt/~nuno/eps/old/2011-12/aula08/Carlson%20\(1996\).pdf](http://www.eq.uc.pt/~nuno/eps/old/2011-12/aula08/Carlson%20(1996).pdf), 1996.
- [63] Aspentech, Aspen Plus, Aspen Plus IGCC Model, USA: Aspen Technology, 2008.
- [64] O. Maurstad, An overview of coal based integrated gasification combined cycle (IGCC) technology, Available 06.2012 from [http://sequestration.mit.edu/pdf/LFEE\\_2005-002\\_WP.pdf](http://sequestration.mit.edu/pdf/LFEE_2005-002_WP.pdf), 2005.

- [65] M. J. Moran and H. N. Shapiro, Fundamentals of Engineering Thermodynamics, USA: Fifth Edition, 2006.
- [66] A. J. Minchener, Coal gasification for advanced power generation, Churchdown: IEA Clean Coal Centre, 2005.
- [67] A.-G. Collot, Matching gasification technologies to coal properties, London: International Energy Agency-Clean Coal Centre, 2005.
- [68] J. Phillips, Different Types of Gasifiers and Their Integration with Gas Turbines, Charlotte: EPRI Advanced Coal Generation, 2004.
- [69] ECN, Coal, Illinois #6 bituminous (#2851), Available 19.09.2013 from <https://www.ecn.nl/phyllis2/Biomass/View/2851>, 2013.
- [70] USDoE National Energy Technology Laboratory, Wabash River Coal Gasification, Available May 2013 from [http://www.netl.doe.gov/technologies/coalpower/cctc/cctdp/bibliography/demonstration/pdfs/wabsh/Final%20\\_Report.pdf](http://www.netl.doe.gov/technologies/coalpower/cctc/cctdp/bibliography/demonstration/pdfs/wabsh/Final%20_Report.pdf), 2000.
- [71] USDoE, KRW Gasifier IGCC Base Cases, Available May 2013 from [http://www.netl.doe.gov/technologies/coalpower/gasification/pubs/pdf/system/krw3x\\_20.pdf](http://www.netl.doe.gov/technologies/coalpower/gasification/pubs/pdf/system/krw3x_20.pdf), 2000.
- [72] J. G. Speight, Gasification Processes, The Chemistry and Technology of Coal, Available June 2012 from CRC net Base, Third Edition, 2012.
- [73] S. Lee, Gasification of Coal, Handbook of alternative fuel technologies, Taylor & Francis Group, 2007.
- [74] D. E. & C. C. Ltd., Davy Process Technology and Johnson Matthey announce Successful Start-up of Datang Phase 1 SNG plant in China, London: Available September 2013 from <http://www.davyprotech.com/pdfs/Datang%20Phase%201%20SNG%20startup%20-%20Press%20Release.pdf>, 2012.
- [75] USDoE, Utah Coal and Biomass Fueled Pilot Plant Kanab, Available November 2012 from [http://www.netl.doe.gov/publications/others/nepa/Utah\\_Biomass\\_Coal\\_FEA\\_1221111.pdf](http://www.netl.doe.gov/publications/others/nepa/Utah_Biomass_Coal_FEA_1221111.pdf), 2011.
- [76] CNET News, Clean-tech startup GreatPoint scales up in China, Available [http://news.cnet.com/8301-11386\\_3-57381270-76/clean-tech-startup-greatpoint-scales-up-in-china/](http://news.cnet.com/8301-11386_3-57381270-76/clean-tech-startup-greatpoint-scales-up-in-china/), 2012.



- [77] Great point energy, News and events, Available 23.09.2012 from <http://www.greatpointenergy.com/newsandevents.php>, 2012.
- [78] Boston Globe and Great point Energy, Massachusetts firm in \$1.25b deal in China, Available 28.10.2012 from <http://bostonglobe.com/business/2012/05/20/mass-firm-deal-china/KBOUYJyGg4r3MTAv65jm1K/story.html>, 2012.
- [79] USDoE, Catalytic gasification, Available 10.12.2012 from [http://www.netl.doe.gov/technologies/coalpower/gasification/gasifipedia/4-gasifiers/4-1-4-4\\_catalytic.html](http://www.netl.doe.gov/technologies/coalpower/gasification/gasifipedia/4-gasifiers/4-1-4-4_catalytic.html).
- [80] USDoE, Development of a Hydrogasification Process for Co-Production of Substitute Natural Gas and Electric Power from Western Coals, National Energy Technology Laboratory, 2009.
- [81] I. E. A. C. RESEARCH, METHANATION CATALYSTS, LONDON, ENGLAND, 1980.
- [82] GreatPoint Energy, Bluegas presentation, Available 12.06.2013 from <http://www.canadiancleanpowercoalition.com/pdf/SNG4%20-%20perlman-presentation1.pdf>, 2008.
- [83] Great Point Energy, Our technology - Clear advantages, Available 13.06.2013 from <http://greatpointenergy.com/clearadvantages.php>, 2013.
- [84] Royal Society of Chemistry, Catalysis, Available 13.06.2013 from <http://www.rsc.org/ebooks/archive/free/CL9780851865843/CL9780851865843-00001.pdf>, 1985.
- [85] USDoE, Coal-to-Synthetic Natural Gas Efficiency and Performance, Available 28.09.2012 from [http://www.netl.doe.gov/technologies/coalpower/gasification/gasifipedia/6-apps/6-4-1a1\\_efficiency.html](http://www.netl.doe.gov/technologies/coalpower/gasification/gasifipedia/6-apps/6-4-1a1_efficiency.html), 2012.
- [86] S. F. Hannemann, Coal-to-Synthetic Natural Gas Efficiency and Performance Coal-to-Synthetic Natural Gas Efficiency and Performance, Siemens Fuel Gasification Technology GmbH, 2009.
- [87] J. Pierobon, Low natural gas prices are driving coal gasification out of business, Available 02.07.2012 from <http://theenergycollective.com/jimpierobon/85951/low-natural-gas-prices-are-driving-coal-gasification-out-business>, 2012.
- [88] J. Wernau, Clean coal plant on hold over low natural gas prices, Chicago Tribune Business: Available November 2012 from <http://articles.chicagotribune.com/2012-05-24/business/chi-clean-coal-plant-on-hold-over-low-natural-gas-prices->

20120524\_1\_natural-gas-north-shore-gas-clean-coal-plant, 2012.

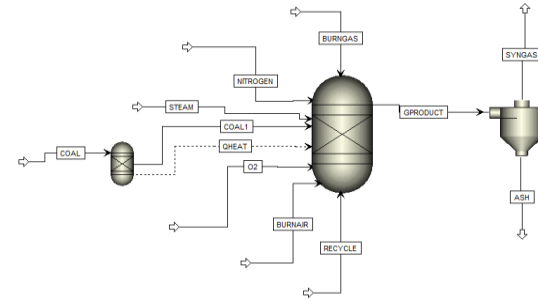
- [89] Sierra Club, Stopping the Coal Rush, Available 10.2012 from <http://www.sierraclub.org/environmentallaw/coal/plantlist.aspx>, 2012.
- [90] S. Berdikeeva, China Turns to Natural Gas to Fuel their Economic Growth, Available November 2012 <http://oilprice.com/Energy/Natural-Gas/China-Turn-to-Natural-Gas-to-Fuel-their-Economic-Growth.html>, 2012.
- [91] W. Sethuraman, China's Easing Grip on Gas Opening Door to North America Exports, Available November 2012 from <http://www.bloomberg.com/news/2012-06-04/china-s-easing-grip-on-gas-opening-door-to-north-america-exports.html>, 2012.
- [92] China Gas Map, Project outline, Available 10.2012 from <http://www.chinagasmap.com/theprojects/coal-to-gas.htm>, 2011.
- [93] ZEUS INTELLIGENCE, GLOBAL GASIFICATION PROJECT INVENTORY, Available 09.2012 from <http://www.zeusintel.com/Gasification/GlobalGasificationProjectListing.aspx>, 2012.
- [94] I. Wright and T. Gibbons, Recent developments in gas turbine materials and technology and their implications for syngas firing, USA: Materials Science and Technology Division, Oak Ridge National Laboratory, 2006.
- [95] Y. S. Kim, J. J. Lee, T. S. Kim and J. L. Sohn, Effects of syngas type on the operation and performance of a gas turbine in integrated gasification combined cycle, Republic of Korea: Dept. of Mechanical Engineering Inha University; Center for Next Generation Heat Exchangers, 2010.
- [96] M. J. B. M. K. Denison, Z. Chen, C. L. Senior and A. F. Sarofim, Using Models To Select Operating Conditions for Gasifiers, Salt Lake City: Reaction Engineering International; USDoE.
- [97] USDoE National Energy technology laboratory, Tampa electric Polk Power Station, Tampa, USA: Available May from <http://www.netl.doe.gov/technologies/coalpower/cctc/cctdp/bibliography/demonstration/pdfs/tampa/TampaFinal.pdf>, 2002.
- [98] World Energy Outlook, "IEA," 2011. [Online]. Available: <http://www.iea.org/publications/freepublications/publication/weo2010.pdf>. [Accessed 10 11 2013].
- [99] Ministry of Energy and resources, Saskatchewan's Strategic Approach to Carbon Dioxide Capture and Storage, Saskatchewan Canada, 2009.

- [100] M. C. Carboa, D. Jansena, J. W. Dijkstra, J. P. v. Buijtenenb and A. H. Verkooijen, Pre-combustion decarbonisation in IGCC: Gas turbine operating window at variable carbon capture ratios, Netherlands: Energy Research Centre of the Netherlands and Delft University of Technology, 2009.
- [101] J. M. Klara and J. E. Plunkett, The potential of advanced technologies to reduce carbon capture costs in future IGCC power plants, U.S. Department of Energy, National Energy Technology Laboratory, 2009.
- [102] ELCOGAS, IGCC PUERTOLLANO; A CLEAN COAL GASIFICATION POWER PLANT, Available  
january 2013 from  
[http://www.ccsnetwork.eu/uploads/publications/ccs\\_network\\_elcogas.pdf](http://www.ccsnetwork.eu/uploads/publications/ccs_network_elcogas.pdf), 2000.
- [103] O. WRONG Maurstad, An Overview of Coal based integrated gasification combined cycle (IGCC) technology, Massachusetts Institute of Technology Laboratory for Energy and the Environment, 2005.
- [104] O. Maurstad, An overview of coal based integrated gasification combined cycle (IGCC) technology, Massachusetts Institute of technology, 2005.

## 8 Appendix

### A) Simulation results of the coal gasification base case

Dry feed entrained flow gasifier results (Shell technical restrictions)



	COAL1	GPRODUCT	NITROGEN	O2	STEAM	SYNGAS
Temperature	25	1690,8	32	32	343	1690,8
Pressure bar	32,04	55,5	5,41	51	51	55,5
Vapor Frac	0,896	1	1	1	1	1
Mole Flow kmol/hr	1825,119	8565,869	741,167	2767	575,3	8565,869
Mass Flow kg/hr	12810,243	194001,479	20796	87212,989	10364,191	194001,479
Volume Flow cum/hr	1291,115	25402,067	3469,809	1339,316	521,811	25402,067
Enthalpy Gcal/hr	1,138	-46,547	0,013	-0,106	-31,901	-46,547
Mole Flow kmol/hr						
H2O	82,454	220,425	0,296		575,3	220,425
N2	51,882	1173,856	735,164	387		1173,856
O2	108,521	trace	4,002	2353		trace
NO2		trace				trace
NO		< 0,001				< 0,001
S	106,164	0,03				0,03
SO2		0,005				0,005
SO3		trace				trace
H2	1475,821	1817,558				1817,558
CL2	0,275	trace				trace
HCL		0,55				0,55
C						
CO		5078,885				5078,885
CO2		138,982				138,982
H2S		94,316				94,316
CH4		0,364				0,364
H3N		0,38				0,38
AR		28,705	1,705	27		28,705
COS		11,813				11,813

Fixed bed gasifier results (BGL technical restrictions)

	BURNAIR	BURNGAS	COAL1	GPRODUCT	NITROGEN	O2	RECYCLE	STEAM	SYNGAS
Temperature F	60	60	77	1810,1	60	392	100	680	1810,1
Pressure psia	500	500	464,7	804,959	500	517	500	510	804,959
Vapor Frac	1	1	0,779	1	1	1	0	1	1
Mole Flow lbmol/hr	108,63	8,55	3044,029	8295,644	157,389	1502,584	9,66	1618,515	8295,644
Mass Flow lb/hr	3147,018	138,683	25339,157	163207,613	4409	48051	684,886	29158	163207,613
Volume Flow cuft/hr	1192,227	88,904	30105,963	253519,257	1732,768	26625,847	12,803	36478,045	253519,257
Enthalpy Gcal/hr	-0,006	-0,07	-16,069	-67,439	-0,009	0,838	-0,007	-40,509	-67,439
Mole Flow lbmol/hr									
H2O			588,916	918,764			0,03	1618,515	918,764
N2	84,73	0,08	42,682	291,248	157,389	7,496			291,248
O2	22,81		205,382			1495,087			trace
NO2									trace
NO									trace
S			74,576						trace
SO2									trace
SO3									trace
H2			2128,527	2593,607					2593,607
CL2			3,947	trace					trace
HCL				7,893					7,893
C									
CO				3291,052					3291,052
CO2		0,02		721,112			0,99		721,112
H2S				71,036			0,46		71,036
CH4		8,45		393,456					393,456
H3N				2,258					2,258
AR	1,09			1,09					1,09
COS				4,06			0,06		4,06
BENZENE				trace			7,81		trace
PROPA-01				< 0,001			0,05		< 0,001
ISOBU-01				trace			0,05		trace
ETHAN-01				0,068			0,21		0,068

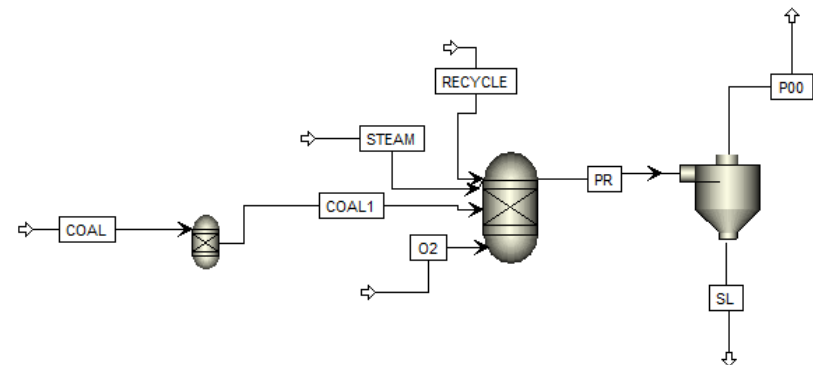
Fluidized bed gasifier (KWR technical restrictions)

	Air	COAL1	GPRODUCT	STEAM	SYNGAS
Temperature C	32	25	1005,5	343	1005,5
Pressure bar	51	32,04	55,5	51	55,5
Vapor Frac	1	0,779	1	1	1
Mole Flow kmol/hr	14408,261	3793,733	22147,007	1188,059	22147,007
Mass Flow kg/hr	417361,679	31579,856	546141,68	21403,21	546141,68
Volume Flow cum/hr	7064,436	2342,334	42887,378	1077,598	42887,378
Enthalpy Gcal/hr	-0,985	-44,151	-138,033	-65,879	-138,033
Mole Flow kmol/hr					
H2O		733,958	1147,606	1188,059	1147,606
N2	11249,97	53,194	11299,902		11299,902
O2	3017,09	255,964	trace		trace
NO2			trace		trace
NO			trace		trace
S		92,943	trace		trace
SO2			< 0,001		< 0,001
SO3			trace		trace
H2		2652,755	3144,695		3144,695
CL2		4,919	trace		trace
HCL			9,837		9,837
C					
CO			5113,256		5113,256
CO2	6,628		1107,095		1107,095
H2S			86,615		86,615
CH4			90,575		90,575
H3N			6,525		6,525
AR	134,573		134,573		134,573
COS			6,328		6,328

Slurry feed entrained flow gasifier results (E-Gas technical restrictions)

	COAL1	O2	SYNGAS	GPRODUCT	Water
Temperature	25	25	1207,3	1207,3	25
Pressure bar	32,04	55,5	55,5	55,5	55,5
Vapor Frac	0,749	1	1	1	< 0,001
Mole Flow kmol/hr	3131,915	2522,878	10397,291	10397,291	2395,729
Mass Flow kg/hr	27451,556	81119,099	212092,518	212092,518	42634,054
Volume Flow cum/hr	1854,697	1078,458	23273,597	23273,597	42,677
Enthalpy Gcal/hr	-45,351	-0,302	-222,947	-222,947	-157,812
Mass Flow					
H2O	710,011		2009,525	2009,525	2298,342
N2	47,74	36,189	84,744	84,744	
O2	204,239	2419,482	trace	trace	
NO2			trace	trace	
NO			trace	trace	
S	78,954		< 0,001	< 0,001	
SO2			0,001	0,001	
SO3			trace	trace	
H2	2090,378		2994,091	2994,091	
CL2	0,593		trace	trace	
HCL			1,186	1,186	
C					
CO			4058,982	4058,982	
CO2			1091,345	1091,345	
H2S			76,437	76,437	
CH4			8,638	8,638	
H3N			0,54	0,54	
AR		67,208	67,208	67,208	
COS			4,596	4,596	

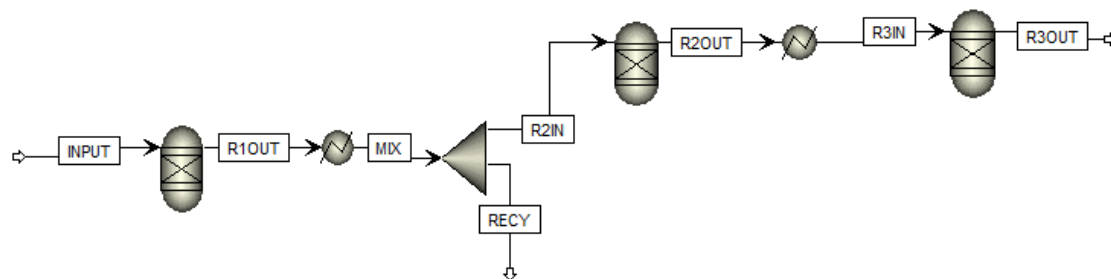
## B) Simulation results of the hydromethanation base case



	COAL	COAL1	O2	P00	PR	RECYCLE	SL	STEAM
Temperature		25	25	600	600	340		343
Pressure bar	51	32,04	51	50	50	51		51
Vapor Frac		0,783	1	1	1	0,394		1
Mole Flow kmol/hr	0	120,599	23,512	339,148	339,148	94	0	200
Mass Flow kg/hr	0	1003,893	755,987	7348,904	7348,904	915,182	0	3603,056
Volume Flow cum/hr	0	74,555	10,97	492,34	492,34	37,86	0	178,957
Enthalpy Gcal/hr		-1,475	-0,003	-14,63	-14,63	-0,018		-11,108
Mole Flow kmol/hr								
H2O		23,332		147,002	147,002			200
N2		1,691	0,337	1,995	1,995			
O2		8,137	22,548					
NO2								
NO								
S		2,955						
SO2								
SO3								
H2		84,329		34,068	34,068	31		
CL2		0,156						
HCL				0,313	0,313			
C						57		
CO				6,167	6,167	6		
CO2				68,753	68,753			
H2S				2,927	2,927			
CH4				77,203	77,203			
H3N				0,067	0,067			
AR			0,626	0,626	0,626			
COS				0,028	0,028			



### C) Simulation results of the methanation base case

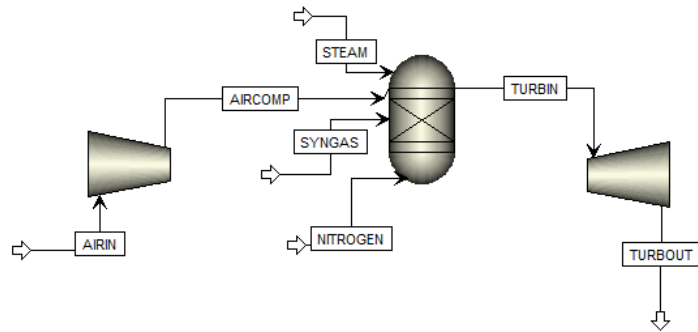


	INPUT	MIX	R1OUT	R2IN	R2OUT	R3IN	R3OUT	RECY
Temperature C	290	281	600	281	438	210	303	281
Pressure bar	27,2	27,1	27,2	27,1	27	27	26,9	27,1
Vapor Frac	1	1	1	1	1	1	1	1
Mole Flow kmol/hr	30089,329	26437,08	26437,08	7138,011	6618,964	6618,964	6453,712	19299,068
Mass Flow kg/hr	427038,268	427038,268	427038,268	115300,332	115300,332	115300,332	115300,332	311737,935
Volume Flow cum/hr	51851,436	44488,946	70762,629	12012,015	14416,185	9509,285	11253,157	32476,93
Enthalpy Gcal/hr	-638,226	-727,356	-637,064	-196,386	-197,064	-212,976	-210,35	-530,97
Mole Flow kmol/hr								
H2O	5506,347	8228,144	8228,144	2221,599	2661,939	2661,939	2824,102	6006,545
CH4	8244,476	10070,601	10070,601	2719,062	2978,586	2978,586	3061,212	7351,539
CO	1233,662	303,209	303,209	81,867	3,159	3,159	0,07	221,343
CO2	2196,521	1300,849	1300,849	351,229	170,413	170,413	90,876	949,62
H2	11433,945	5059,899	5059,899	1366,173	406,786	406,786	79,37	3693,726
N2	1474,377	1474,377	1474,377	398,082	398,082	398,082	398,082	1076,295





## F) Simulation results of the gas turbine base case



	AIRCOMP	AIRIN	NITROGEN	STEAM	SYNGAS	TURBIN	TURBOUT
Temperature C	438	15	93	246	45	1338	741
Pressure bar	18	1	27	32	37	2	0
Vapor Frac	1	1	1	1	1	1	1
Mole Flow kmol/hr	110253	110253	16375	1705	21155	141839	141839
Mass Flow kg/hr	3181420	3181420	459474	30712	417634	4089240	4089240
Volume Flow cum/hr	404824	2639480	18849	2048	12308	11878400	117666000
Mole Flow kmol/hr							
AR	1014	1014	38		167	1219	1219
CH4					11		
CO					10030		
CO2	33	33			264	10337	10337
H2					5270		
N2	85248	85248	16244		1007	102499	102499
O2	22866	22866	88			15284	15284
H2O	1092	1092	5	1705	4407	12498	12498



